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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

REMEDIAL ACTIONS IN CASE OF GROUNDWATER CONTAMINATION OF SANDY AQUIFERS (1966)

Final Report

by

L.H. BAETSLE (CEN)
W. de LAGUNA (Oak Ridge National Laboratory)
J. SOUFFRIAU (BelgoNucléaire)

1968

Report prepared by CEN
Centre d'Etude de l'Energie Nucléaire, Mol - Belgium

Euratom Contract No. 010-65-1 WASB

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Printed by Guyot
Brussels, December 1968

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SUMMARY

Hydrogeologic investigations on the SCK-CEN site at Mol, Belgium, has shown that the water table aquifer is made up of two sandy formations with different permeability and partly sealed from the underlying aquifer by a succession of tiny impermeable layers.

The removal of non adsorbed radioactive contamination from the underground by pumping, called "scavenging", follows the classical pattern of pumping from a water table under the condition that the two layers with permeabilities 1.12 cm/min. and 0.56 cm/min. and their respective thicknesses are considered. By injecting HF and $K_2H_2Sb_2O_7$ into the aquifer a chemical barrier is formed which is specific for Sr tests on the penetration of reagents tagged with ^{82}Br into the aquifer have indicated that 1.5 to 2 void volumes are sufficient in order to replace all the interstitial fluid by reagent. Zones with different permeability have to be treated separately. The delay coefficient of the chemical barrier installed in the water table aquifer at Mol amounts to 20, i.e. the Sr moves twenty times slower than the groundwater.

KEYWORDS

REACTOR SITES
BELGIUM
HYDROLOGY
GEOLOGY
SAND
DEPOSITS
POROSITY
LAYERS
DECONTAMINATION
INJECTION
HYDROFLUORIC ACID
ANTIMONY OXIDES
BROMINE 82
TRACER TECHNIQUES
GROUNDWATER
DIFFUSION
ZONES
CONTAMINATION

C O N T E N T S

REMEDIAL ACTIONS IN CASE OF GROUNDWATER CONTAMINATION OF SANDY AQUIFERS

1. PURPOSE AND SCOPE OF STUDY

The purpose of the studies described below was to explore the practical possibilities of remedial action in the very unlikely event of serious contamination of the groundwater at Mol, Belgium, resulting from an accident at one of the nuclear facilities. These facilities include three experimental reactors, a radioactive waste treatment plant, and, chemical plant for the reprocessing of nuclear fuel. This study relies on the assumption that a big accident in these facilities might release potentially disastrous quantities of radioactive materials into the groundwater. There is general agreement that the probability of such an accident is very small.

Following the description of the environment, as determined by test drilling and a series of rather specialized hydrologic tests in the field, an attempt is made to assess the practical potential of the two countermeasures which have been considered for possible use in the event of radioactive contamination of the groundwater : "scavenging" and installation of chemical barriers to reduce the migration rate of the critical contaminant extraction of the radioactivity from the aquifer by pumping called. These two countermeasures are described in detail in later sections of this report.

2. STRATIGRAPHY AND HYDROLOGY AT THE CEN TEST SITE

In the summer of 1965, when plans were being made for the tests of "scavenging" and chemical barriers, as described below, a decision was reached to move the preexisting site of the field experiments from the property of Eurochemic to the land occupied by CEN at the eastern end of the site [1] [2]. The first well drilled provided samples to determine the local stratigraphy and was later used as the pumped well for the scavenging tests.

The log of the well is as follows : drilled at the CEN site, October 4 to 11, 1965; bailed in to 28 meters, then cores removed, in order to sink 200-mm casing to final depth of 40 meters.

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*) Manuscript received on 22 August, 1968.

Pleistocene Cover - 0 to 1.5 meters. Poorly sorted yellow sand with a few well-rounded pebbles at 1.5 meters.

Mol Sand - 1.5 to 9.0 meters. Medium-grained sand, fairly well sorted, no glauconite or lignite visible. Color, yellow to 4.0 meters; deeper, very light gray, almost white. Ten to fifteen layers of tough white clay, 2 to 3 mm thick, between 5 and 9 meters.

Kasterlee Sand - 9.0 to 25.0 meters. Fine- to medium-grained, well-sorted sand; color, very light gray. No clay layers and very little interstitial clay to 20 meters; from 20 to 25 meters, slightly more interstitial clay and color slightly darker, but difference is believed insignificant.

Kasterlee-Diest Transition - 25 to 28 meters. Medium-fine sand as above, but content of fine-grained glauconite and gray clay increases slowly with depth in this zone. Color grades downward from light green to medium green. This sand is part of the Kasterlee,containing, with depth, increasing amounts of material reworked from the underlying Diest. Although probably slightly less permeable than the main body of the Kasterlee, it will be considered part of the Kasterlee hydrologic unit.

Diest Sand - 28.0 to 40 meters. Medium-fine sand, 25 to 50% glauconite, pores largely filled with fine-grained glauconite and gray clay. Several thin brown-gray clay layers between 28 and 31.5 meters with total thickness of about 10 cm. At 35 meters general color changes from bright green to brownish green due to presence of brown clay mixed with the sand. All is very poorly permeable compared to overlying beds.

The Mol-Kasterlee contact was set with confidence at a depth of 9 meters in this well on the basis of mechanical analysis of samples which showed a change identical with that found 5.5 meters deeper in the 1961 Eurochemic well. The Kasterlee-Diest contact was set at 28 meters with confidence on the basis of the abrupt marked increase in coarse-grained glauconite and clay. This contact was 5 meters deeper at Eurochemic. The clay layer at this contact, which consisted of at least 12 cm of solid clay at Eurochemic, was represented in the CEN well by a number of thin clay layers, but there was enough interstitial clay and fine-grained glauconite to reduce greatly the permeability of the upper part of the Diest penetrated by the test well.

These several investigations complete the description of the geohydrologic environment at Mol and guarantee a sufficient precision for the design of field tests [3].

The geohydrologic of the CEN test site at Mol may be summarized as follows :

The "water-table" aquifer consists of two sand: the upper, the Mol sand, is relatively coarser and less well-sorted; the lower, the Kasterlee sand, is somewhat finer grained and very well sorted. Although the two might well be treated as a single hydrologic unit in studies of water supply, they probably differ sufficiently in permeability and in hydrologic homogeneity so that they should be treated as separate units in studies involving the movement of contaminated water through the aquifer (Fig. 1).

In the bottom 4 meters of the Mol sand there are a number of thin clay layers. While these hardly prevent, they certainly impede the movement of water between the Mol sand and the Kasterlee sand. This is equivalent to saying that in the lower half of the Mol sand the vertical permeability is probably greatly reduced by the thin clay layers.

3.SCAVENGING AND BARRIER BUILDING

Having described in a general way the geohydrological environment at Mol, we can look more closely at the possible countermeasures which might be attempted in the very unlikely event of serious contamination of the groundwater. Depending on the location of the accident, the contaminated groundwater might need to travel only a few tens of meters to reach the Maas-Schelde canal, or the sand pits and interior canal which drain into it, or it might need to travel underground up to 3 km if it were moving south toward the Mol Nete River. The rate of movement of the groundwater is known to be of the order of 5 to 10 cm per day; and, where the pH of the groundwater is less than 4.0 or 4.5, ^{90}Sr has been shown to move at substantially the same rate as the groundwater. In the event of an accident occurring some 50 meters from one of the lakes draining into the barge canal, there would be an interval of 500 to 1000 days before the canal became contaminated. A year and a half to 3 years is long enough to make possible even fairly complex remedial action if the plan of action has been decided on in advance. On the

other hand, if the groundwater was contaminated at a location from which it must flow some 3 km to reach the Mol Nete, the journey would require roughly 100 years, a long time by many standards, but only some three to four half-lives for ^{90}Sr. This would be sufficient to reduce appreciably the concentrations of ^{90}Sr, but not enough to ensure decay to safe levels.

These are essentially the extreme examples; and, although many additional factors would have to be considered, they do suggest that, in the event of accidental groundwater contamination, there will be time for remedial action before the contamination reached flowing surface water, where virtually all control over it would be lost, but not enough time to reduce the ^{90}Sr to safe levels by natural decay and dilution.

When a contamination has occurred which is potentially dangerous for the surrounding population there are only two possible remedial actions to take :

1) the contamination is removed from the aquifer by pumping from one or more wells and the water is returned to the environment after proper decontamination;

2) the contamination is confined within a zone where it does not provide a significant hazard and is kept in this area for a period of time long enough to let the critical radionuclide decay.

The first method is called "scavenging" of the contaminated aquifer, the second measure relies on the reduction of the migration rate of certain radionuclides by introduction of ion exchange materials into the aquifer or "chemical barriers"

3.1 Scavenging of underground contamination

Scavenging consists in sinking one or more wells in or near the body of contaminated groundwater and pumping it out, hence the name scavenging. The contaminated pump effluent is either processed to remove the activity and the then harmless water returned to the environment.

The feasibility of scavenging as a remedial measure at any site depends in large measure on the volume of water that would have to be pumped out of the ground to clean up the results of an accident; that is, on the rate of pumping and duration of pumping that would be required. In a homogeneous but not necessarily isotropic aquifer, and by this is meant an aquifer in

which the horizontal permeability is the same from top to bottom, but in which the vertical permeability may differ from the horizontal, the purely hydrologic problem of determining the required minimum pumping rate and the total volume to be pumped will present no difficulties. A more serious problem will be to make the proper allowance for the additional volumes that will have to be pumped because the contaminants move less rapidly than the water due to their adsorption on minerals in the aquifer.

The purely hydrologic problem becomes appreciably more difficult if the aquifer consists of layers with different permeabilities. These "scavenging" tests at Mol were mainly tests to determine the relative permeability of the Mol sand and the Kasterlee sand and to evaluate, at least qualitatively, the hydrologic importance of the clay layers in the lower half of the Mol sand.

3.1.1 Scavenging tests

After the 200-mm casing of the test well at the CEN site had been sunk to 40 meters, it was jacked back in stages to 28 meters, that is to the Kasterlee-Diest contact, and the open hole back-filled to this level with clay. A 150 mm plastic well screen, 28 meters long, was then lowered inside the casing, the annulus filled with pea gravel, and the outer casing removed, leaving a gravel-packed well, even though the gravel packing was somewhat thin. The well as pumped through a drop pipe that extended nearly to the bottom of the screen, so that the full saturated thickness of the Mol and Kasterlee sands was subject to essentially the same pumping head. An electrically driven centrifugal pump was used, and the discharge was carried by light tubing to the edge of the old sand pit about 200 meters to the southeast. Six plastic observation wells were installed in pairs at distances from the pumped well of 5, 10 and 15 meters. One well of each pair was "shallow", and was screened from 4.0 to 4.5 meters deep in the Mol sand just above the thin clay layers. One well of each pair was "deep", and was screened from 18.0 to 18.5 meters deep, near the middle of the Kasterlee sand. Later, a seventh well was installed, 10 meters from the pumped well and screened from 6.0 to 6.5 meters deep, about in the middle of the lower part of the Mol sand which contains the thin clay layers (Fig. 2).

Four test pumpings were made during May and June 1966. On each occasion a chemical tracer was added to one of the observation wells and its arrival in the pumped well detected by periodic measurement of the electrical resistance of the water in the pump discharge. The tracer used was a mixture of ammonium chloride, methanol and water, in such proportions that the mixture has essentially the same density as the groundwater. In the first test, during which about 500 m^3 of water was pumped, the tracer consisted of 8 kilos of ammonium chloride, 20 liters of methanol, and 32 liters of water. In the fourth test, during which 4840 m^3 of water was pumped, the tracer consisted of 50 kilos of ammonium chloride, 100 liters of methanol and 200 liters of water. This represents an average concentration of chloride ion in the pump discharge of about 10 parts per million, the peak concentrations were several times the average. The constant temperature of the groundwater makes it possible to measure these changes in concentration with confidence.

For the purposes of the following discussion, we will consider that the water-table aquifer is actually composed of two separate aquifers. The upper is the saturated portion of the Mol sand, which extends from 3 meters below the land surface to a depth of 9 meters, and so is 6 meters thick. The lower is the Kasterlee sand which here extends from 9 meters to 28 meters and hence is 19 meters thick. We will assume that during the several tests water flowed radially in towards the pumped well and that, although the drawdown was greater in the lower aquifer than in the upper, only a negligible amount of water moved down from the upper aquifer into the lower. This is equivalent to assuming that, during the short time required for one of these tests, the thin clay layers in the lower part of the Mol sand were effectively impermeable. The longest test lasted only for 220 h , about 9 days and the difference in head across the clay layers, 10 meters from the pumped well, was only a little over 0.1 meter.

This assumption, that the Mol and Kasterlee sands may be considered as separate aquifers, at least over limited periods of time, is so vital to the whole hydrologic study here described that a short additional test was made to investigate the question in more detail. To this end, a seventh observation well was installed 10 meters from the pumped well and screened from 6.0 to 6.5 meters below the surface; that is, about in the middle of that lower part of the Mol sand which contains the thin clay layers.

During a preliminary test the main well was pumped for a little over 8 h and the drawdown recorded in all three wells 10 meters from the pumped well. Figure 3 shows the results plotted on rectangular coordinates. There is slightly more drawdown in the 6.5 meter deep well (10M) than in well 10S, but the plots are very similar and differ markedly from the plot of the drawdown in the 18.5 meter deep well (10D). Of considerable interest is the almost instantaneous drawdown of 0.2 to 0.3 meter in well 10D when pumping started, a feature characteristic of an artesian as opposed to a water-table aquifer. In any case, this test tends to confirm the validity of dividing the "water-table aquifer" into two separate aquifers.

A vertical cylinder of the upper aquifer 6 meters high, with a radius of 5 meters and with a porosity of 35%, contains 165 m^3 of water. However, to pump out this much water from the upper aquifer, it was necessary to pump 352 m^3 from the well, as both the upper and lower aquifers contributed water. Therefore, the lower aquifer contributed 187 m^3, while the upper aquifer was contributing 165 m^3. The upper aquifer is 6 meters thick, the lower 19. The ratio of their permeabilities is consequently the ratio of 165 divided by 6 to 187 divided by 19, or 27.4 over 9.85 or 2.78. The pumping rate from the upper aquifer was 165 m^3 divided by 16.0 h , or 10.3 m^3 per hour. The pumping rate from the lower aquifer was 187 m^3 divided by 16.0 h or 11.7 m^3 per hour. The upper aquifer provided 46% of the water, while the lower provided 54%.

In the first test the tracer was added to the shallow well, 5 meters from the pumped well (well 5S). The pumping rate averaged 22 m^3 per hour. The tracer was first detected after 10.5 h or after 231 m^3 of water had been pumped. The average travel time, amounted to 16.0 h or 352 m^3. Figure 4 shows the break-through curve of the chemical tracer.

In the second test the tracer was added to well 5D; 5 meters from the pumped well and screened between depths of 18.0 and 18.5 meters. The average puming rate was 21 m^3 per hour. The average arrival time for the tracer was 45 h , during which time 945 m^3 was pumped (Fig. 5). A saturated cylinder of sand with a radius of 5 meters, a height of 19 meters and a porosity of 35% contains 522 m^3 of water. By calculations similar to those above, the ratio of the average permeability of the sand in the lower can be shown to be 2.56 . The pumping rate from the

upper aquifer was 9.4 m^3 per hour and from the lower aquifer 11.6 m^3 per hour. This time the upper aquifer provided 45% of the water and the lower aquifer 55%.

In the third test the tracer was introduced into well 10S. The average arrival time of the tracer in the pumped well was 78 h , or after 1560 m^3 of water had been pumped (Fig. 6). The average pumping rate for this test was 20 m^3 per hour. The ratio of the average permeability of the upper aquifer to the lower was 2.32. The upper aquifer at a pumping rate of 8.5 m^3 per hour supplied 42.4% of the water, while the lower aquifer, at a pumping rate of 11.5 m^3 per hour supplied 57.6%.

In the fourth and final test of the series the tracer was introduced into well 10D. The average arrival time of the tracer in the pumped well was 185 h or after 4070 m^3 of water had been pumped. The average pumping rate for this test was 22 m^3 per hour. The ratio of the average permeability of the upper aquifer to the lower was 2.75. The upper aquifer at a pumping rate of 9.75 m^3 per hour supplied 46.5% of the water, while the lower aquifer, at a pumping rate of 12.25 m^3 per hour, supplied 53.5% (Fig. 7).

The ratio of permeabilities in the Mol and Kasterlee aquifers ranged from 2.78 to 2.22, and the percent of total water furnished by the upper aquifer ranged from 47% to 42.4%. This relatively small variation in results, which depend, in part, on the validity of the assumptions used for their calculation, suggests that the assumptions are justified for the conditions of the test.

During the first test, the one in which the tracer was introduced into well 5S, the changes in water level with time were measured at frequent intervals in all six observation wells then available. This made it possible to calculate the transmissibility, permeability and storage coefficients for both the upper and the lower parts of the water-table aquifer.

Figure 8 shows the drawdown observed in the shallow observation well located 5 meters from the pumped well. The drawdown is plotted in meters on a linear scale, and the time, in hours, is plotted on a logarithmic scale, to permit determination of the transmissibility, permeability and storage coefficient by use of the "Jacob" method [4]. When the points representing

the individual water-level readings in the observation well are so plotted, in general it will be found that after a certain time they fall on a straight line. In the cas of Figure 8 the readings taken prior to about 3 h of pumping do not fall on this line and should be disregarded. Two values are obtained from the straight line drawn through the later points. One is the intercept of this line, extended, with the time axis called "t_o" (0.16 h in Fig. 8) and the other is the slope of the line represented by the observed drawdown during one log cycle on the time scale, called "Δh" (0.46 meters in Fig. 8).

The first quantity to be determined is "T" or the transmissibility of the aquifer. In the metric system, this may be defined as the number of cubic meters of water per hour that would flow through each meter width of the aquifer measured normal to the gradient, if the gradient in the aquifer were unity. This is equivalent to the permeability of the aquifer multiplied by the aquifer thickness.

In the "Jacob" method, the transmissibility is given by $T = \frac{0.183\ Q}{\Delta h}$, where "Q" is the rate of discharge of the pump in cubic meters per hour. As we have seen, the discharge of the pump for the upper part of the aquifer during the first tracer test was 10.3 m^3 per hour and Δh is 0.46 meters (see Fig. 8). The transmissibility of the upper part of the aquifer is, therefore 4.10 m^2 per hour. The thickness of this part of the aquifer (the Mol sand) is 6 meters, so that the permeability is 4.10 m^2 per hour divided by 6 meters, or 0.683 meter per hour, or 1.14 cm per minute.

The storage coefficient may be defined as the volume of water released from storage per unit surface area of aquifer per unit change in the component of head normal to that surface. In the metric system, for a water-table aquifer, this may be defined as that fraction of a cubic meter of water that would be released by draining a cubic meter of saturated sand. An important,and to some extent unrealistic, part of the assumption is that all of the water that will come out of the sand assumed to drain out immediately, whereas in fact some drainage continues for an appreciable time. This is particularly true of a semi-artesian aquifer such as the Mol or Kasterlee sands. The storage coefficient, "S", in any units, is a dimensionless number and in the present case is given by :

$$S = \frac{2.25 \times T \times t_o}{r^2}$$

where

"T" is the transmissibility,

"t_o" is the intercept of the straight line with the time axis,

"r" is the distance of the observation well from the pumped well, in meters.

For the case in question

$$S = \frac{2.25 \times 4.10\ m^2/h \times 0.16\ h}{(5m)^2} = 0.059$$

For the drawdown data from well 10S, shown in Figure 9, $\Delta h = 0.47$ m, $t_o = 0.7$ h , r = 10 meters and Q, the pumping rate, remains 10.30 m^3 per hour. Using the same formulas, T = 4.01 m^2 per hour, the permeability is 1.115 cm per minute, and S = 0.0631. For the drawdown data from well 15S, shown in Figure 10, $\Delta h = 0.43$ m, $t_o = 1.4$ h , T = 4.38 m^2 per hour, the permeability is 1.22 cm per minute and S = 0.0614.

The procedure is the same for the analysis of the drawdown data from the three "deep" observation wells, shown in Figures 11, 12 and 13. The pumping rate, "Q", for this aquifer, however, was found to be 11.70 m^3 per hour, and T, the transmissibility must be divided by 19 meters, the thickness of the lower (Kasterlee sand) aquifer to get the permeability. So calculated, the data from well 5D gives for "T", the transmissibility, 6.12 m^2 per hour, a permeability of 0.552 cm per minute, and a storage coefficient of 0.0148. The data from well 15D gives a transmissibility of 6.8 m^2 per hour, a permeability of 0.596 cm per minute and a specific yield of 0.0156. These results are summarized in Table I.

As a first approximation and as a rough check on the validity of the various assumptions on which these calculations are based, it is of interest to compare the relative proportions of the water pumped from the Mol sand and the Kasterlee sand with their relative transmissibilities, as determined by the

pumping test. From the four tracer tests the Mol sand appeared to furnish an average of 45% of the water pumped, while the Kasterlee furnished 55%. The average transmissibility of the Mol sand was calculated to be 4.16 m^2 per hour, that of the Kasterlee 6.41 m^2 per hour, giving a combined transmissibility of 10.67 m^2 per hour. The Mol sand contributes about 40% of this transmissibility and should, therefore, contribute 40% of the water, not 45% as determined by the average of the tracer tests. Similarly, the Kasterlee contributes 60% of the total transmissibility, but only 55% of the water, as measured by the tracer tests. Although closer agreement of these values would be a matter of satisfaction, so many assumptions, which are at best only approximations, must be made in order to carry out the calculations, that even this rough agreement gives some assurance that, for example, we are justified in subdividing the water-table aquifer into the Mol sand and the Kasterlee, and treating these as essentially independent aquifers, where short periods of time are involved. The agreement, although rough, also suggests that the permeabilities and storage coefficients as calculated, are very probably substantially correct.

Having determined these properties, the next question is, what conclusions, of practical interest, may we deduce from them. One obvious use to make of them is to calculate the probable rate of movement of the groundwater. Over much of the area of interest, the water-table gradients are of the order of one part in 500. The groundwater velocity (V)in the Mol sand would be, therefore (taking the porosity as 35%),

V (Mol sand) = 1.16 cm/min x 1/500 x 1/0.35 x 1440 min/day = 9.4 cm per day,

which is about the velocity measured over short distances with various tracers. In the Kasterlee, the velocity will be reduced in proportion to the permeability.

V (Kasterlee sand) = 9.4 cm/day x 0.562/1.16 = 4.7 cm/day.

.1.2 Design of a scavenging system

If the hypothetical accident was known to have released a liquid contaminant in an area roughly 20 meters in diameter, the results of the third chemical tracer test can be applied directly. There is some measure of assurance that the contamination would, for a period of weeks, months or even a year, be largely confined to the Mol sand. A well, 9 meters deep installed at the center of the

contaminated area, could then be pumped at any convenient rate, say 2 m^3 per hour, until a total of 780 m^3 of water had been removed from the Mol sand. This volume would not only remove the water in a cylinder of the Mol sand with a radius of 10 meters, which is 660 m^3, but would also be sufficient to make allowance for the hydrodynamic dispersion, as represented by the "trailing edge" of the tracer, which came through with the pumped water after the peak concentration of the tracer had passed. This raises the question as to whether or not the dispersion of the chemical tracer is representative of the dispersion of the water and, more importantly, whether it represents the dispersion to be expected of ^{90}Sr, which is slightly adsorbed by the sand, the amount of the adsorption depending on the pH of the water. However, the pH of the groundwater at Mol is sufficiently low (about 4) so that adsorption of the strontium is not appreciable and that the results obtained with the chemical tracer may be applied directly.

This proposed design of the scavenging well and the prediction of the volumes that would need to be pumped are for a "minimum" accident, as measured in terms of the area that is assumed to be contaminated. If the groundwater under a larger area must be cleaned up by scavenging, other factors should be considered and more sophisticated methods employed.

For simplicity, we might, however, begin with a very rough calculation of what could be done with a single well penetrating the full thickness of a homogeneous water-table aquifer, pumped at a constant rate.

Under the conditions existing at the Mol site the surface area within the hypothetical parabolic cone of influence created by continuous pumping on a fully screened well at 20 m^3 per hour would cover about 172,000 m^2 and this area of the aquifer would contain about 1,370,000 m^3 of water. This would require 68,500 h , or nearly 8 years, to pump out, if the flow rate were the same along all the flow lines. However, this is not the case,and about all one can say is that, depending on the exact area contaminated, the pumping time might well be measured in decades.

A simple example of a more flexible system would consist of a row of wells fully penetrating both sections of the water-table aquifer and each pumped at a rate of about 2.2 m^3 per hour. If 10 such wells were spaced 30 meters apart, they would form a hydrologic barrier 300 meters long, to which we can apply directly the

results of our field tests. The drawdown around a pumping well is directly proportiona to the rate of pumping so that we can use the plots of the data from the pumping test by dividing the vertical scale by 10 (Fig. 14). The effects of pumping for a longer period of time may be obtained by extending to the right, over several more log cycles, the straight lines obtained by the semilogarithmic plots of the field test data. Thus we see that after 1 year of pumping, the drawdown in a shallow observation well, 15 meters from the pumped well, will be 0.164 meter and after 10 years will be 0.208 meter. For a deeper (18.5 meters) observation well at the same distance, the equivalent drawdowns will be 0.144 meter and 0.175 meter. If the observation well is located half way between two pumped wells, 30 meters apart, the total drawdown will be just twice the values given, for drawdowns are directly additive. This would appear to offer more than enough drawdown to ensure a gradient toward each of the pumped wells sufficient so that no contaminated water could pass between any two of them. In practice, two or more observation wells would be screened at different depths between each pair of pumped wells and the pumping rate from each well adjusted to the minimum safe value.

The volume that would have to be pumped may be calculated on the assumption that half of the water comes from each side of the barrier. If the contaminated area is contained within a square area 300 meters on a side, the total volume of water contained, assuming now the actual thickness and porosity of the water-table aquifer at Mol, will be 300 meters x 300 meters x 25 meters x 35%, or about 800,000 m^3. Since an equal volume of water will have to be pumped from the other side of the barrier, the total to be pumped will be 1,600,000 m^3 which at a rate 22 m^3 per hour (from the 10 wells) will require about 82,000 h , or nearly 10 years. At first glance this would appear to offer little advantage over the use of a single larger well, as described above, but in practice the large margins of safety could be reduced on the basis of field observations, and the pumping rate or pumping time considerably reduced.

Chemical barriers

A chemical barrier is built into the ground by injecting chemical reagents which react with the silica and increase its adsorptive capacity for a given nuclide. Since Sr is the critical contaminant in the Mol area and since extensive reseach

was carried out on the recovery of Sr from fission product by inorganic ion exchangers the efforts were limited to this hazardous radionuclide.

3.2.1 Preliminary laboratory experiments

A series of laboratory tests with a variety of chemical reagents has shown that both HF and Cp4 [x] may be used as etching reagents and that only the salts of Fe, Sn and Sb increase the adsorptive capacity of the porous medium for Sr. Table II shows the most promising results.

It is striking that $K_2H_2Sb_2O_7$ is the most interesting water soluble salt used. The use of the etching mixture Cp4 did not provide better results than HF and is much more expensive. The concentration 0.050 M seems to be the lower limit for $K_2H_2Sb_2O_7$ as with 0.025 the uptake capacity is much lower (43 instead of 63).
A systematic study of the influence of the HF and $K_2H_2Sb_2O_7$ concentrations together with the resulting change in permeability of the medium is shown in Table III. It may be concluded that the corrosion of the particle surface by HF is the principal reason for the decrease in permeability.

The best result is obtained with the combination 10% HF and 0.05 M $K_2H_2Sb_2O_7$ because no decrease in permeability was observed while the uptake capacity still attains 53 void volumes.

3.2.2 Field tests on the penetration of reagents

Several injection methods were examined but only one method has proven to be practical : pumping of reagents into a fully screened well.

In order to test the validity of the method a well was drilled up to 16 m depth and a fully screened PVC tube installed. Seven observation "dry holes" each one meter apart were installed along the perpendicular axis. The experiment consisted of slowly injecting water tagged with ^{82}Br into the screened well and periodically halting the injection for taking readings at half-meter intervals in each of the "dry wells" with a gamma-sensitive scintillation probe. The concentration of the tracer was such that when all the water in the pores of the sand adjacent to the dry well had been completely replaced by the injected fluid, the probe recorded from about 4000 to a little over 5000 counts per minute.

(x) Cp4 = 10 vol. HF (40%), 100 vol. HAc, 160 vol. HNO_3 fum , 29 vol. Br_2

Figures 15 and 16 show the results of the test as recorded in the dry hole 1 meter west of the injection well. A cylinder of sand 14 meters high, the distance from the water table to the bottom of the injection well, and 1 meter in radius, with a mean porosity of 38.5% contains 17 m^3 of liquid, or "one pore volume" for a dry well at the distance indicated. After one-half pore volume has been injected, some activity was detected in the dry well down to a depth of 9 meters; that is, down to the Mol-Kasterlee interface. There were marked peaks at depths of 3.5, 6.0 and 8.0 meters, but the counts of 3000 to 3500 per minute showed that even at these depths not all of the original water had been replaced. After one pore volume had been injected, the greater part of the dry well down to 9 meters showed close to 4000 counts per minute, or essentially complete replacement of the original water and one peak had appeared in the Kasterlee at a depth of 14 meters. After two pore volumes, virtually the whole of the dry well read 4000 counts per minute or more; and, while further injection, up to 4.5 pore volumes, increased the readings somewhat, the difference may, in part, be attributed to variations in the response of the probe.

Figures 18 and 19 show the results in the similar dry well, 1 meter east of the injection well. After one-half pore volume of "tagged" water had been injected, some activity was detected in the dry well down to but not below the Mol-Kasterlee contact. After two pore volumes, the Mol sand was saturated with the injected water and a little activity was appearing below 9 meters, in the Kasterlee. However, even after 4.5 pore volumes, all of the Kasterlee was not saturated with the "tagged" water, and an even larger column of reagent would have had to be injected to treat the whole section of the aquifer satisfactorily.

Figure 20 shows the readings made in the dry well, 2 meters east of the injection well. After one-half pore volume (34 m^3) of tagged water had been injected, a few low peaks had appeared down to a depth of 7 meters; and, after one pore volume, most of the original groundwater in the Mol sand had been replaced, except for one "low" at 7 meters. However, no activity had yet reached the dry well through the Kasterlee. The figure also shows the readings made in a dry well, 3 meters from the injection well, after one-half pore volume for a cylinder of this radius, or 76.5 m^3, had been injected. Much, but not all, of the water in the Mol sand had been replaced,and there was a trace of activity just below the Mol-Kasterlee contact. By analogy with the readings in the other

dry wells, we may conclude that, if it had been feasible to inject one pore volume, virtually all of the Mol sand would have been "treated", but that, with a well so screened, four or more pore volumes might well be required to treat the screened section of the Kasterlee as well.

In evaluating these tests, it is important to remember that the screen penetrates two aquifers : 7 meters of saturated Mol sand and 7 meters of Kasterlee. From the pumping test, we determined that the Mol has a permeability of 1.16 cm per minute and the Kasterlee a permeability of 0.562 cm per minute, or a ratio of about 2 to 1. If the same permeabilities are present in the injection test area, then for every "pore volume" injected, 66% went into the Mol and 35% into the Kasterlee. Therefore, in the above discussion, when we say "one pore volume has been injected", we should say "one and one-third pore volumes have been injected into the Mol sand and two-thirds pore volume into the Kasterlee". Clearly, in any attempt to build a real chemical barrier, the Mol and the Kasterlee sands should be injected separately.

The most important conclusions from these test are :

1. The two zones with different permeabilities (coarse sand of Mol and fine Kasterlee sand) are not saturated at the same time as might be expected.
   As a rule the tracer arrives at the same level at each row of tubes after 1.5 to 2 times the void volume.
2. The shape of the zone does not change within 7 days after the injection.
3. Any change in pumping rate between 0.5 and 1 void volume/h per meter distance from the injection well does not influence the shape of the zone.

.3 Design and preliminary tests of a model barrier

The layout of the model barrier, as originally constructed, is shown in Figure 20. Two wells were first installed 3 meters apart and screened from 4.5 to 5.0 meters below ground surface. The water table in this area is 2 meters below ground surface. One of these two wells was for the injection of tracers and the other was pumped with a Moyno-type pump. Half-way between these two wells, on a line normal to a line joining them, three wells were installed for the injection of barrier-building chemicals. One of these was on the line between the tracer-injection and the pumped well, the other two were 0.75-meter distant from the first on either side. These three wells were screened from the ground surface to

a depth of 6 meters. Nine dry wells, or dry holes, 6 meters deep, were located around the barrier-building wells as shown. All these wells were made of plastic tubing.

The sand in this area was unusually rich in lignite; and, although we were tempted to move the barrier to a more representative location, the decision was made to attempt first some preliminary tests.

In one of the first of these, a tracer solution containing both the chemical tracer and tritium was introduced into the tracer-injection well and flushed out into the formation with a few liters of water. The pump was then started and samples collected every hour. The conductivity and tritium content of each sample was determined. The results are shown in Figure 21. The curves are plotted directly in terms of pumping time. The rate of pumping decreased during the first few hours, and a correction was made for this in calculating the volumes pumped for each crest and for each average travel time for the tracers.

The results were unexpected in that they show clearly two crests for each tracer. For the tritium the first crest came after 43 m^3 and for the chemical tracer after 46 m^3. The second crest, which was asymetric, came for the tritium after 73.5 m^3 and the average after 76.5 m^3; for the chemical tracer, after 84 m^3 and 87 m^3.

The volume of a cylinder of sand with a radius of 3 meters and a height of 3 meters is 85 m^3. If the porosity is 35%, the liquid volume contained is about 30 m^3. Even if we assume that the short screen of the pumped well removed all of the water in the cylinder indicated, the first peak came through only after one-third more water than would be expected had been pumped. A possible explanation is that the aquifer is not homogeneous, so that the pumped well draws less water from the direction of the tracer-injection well than from some other direction.

The second and larger peaks raise an even more difficult problem, not only because such a large volume had to be pumped to pull the tracer a distance of only 3 meters, but also because there is no rational explanation for obtaining two peaks. At any one point, it is not possible to have two sets of flow lines going in different directions. The tracer "cloud" around the injection well at the start of the experiment is not a point and we must suppose that, by change, part

of it lay below a hydrologic barrier, such as a thin clay layer, and part above. We must further suppose that the part of the cloud below the barrier followed a longer path to the pumped well, or that the rate of flow was slower along the lower path. Several other tests with the same arrangement of wells gave much the same result, so that the test illustrated in Figure 21 is representative.

One interesting point shown in Figure 22 is that the chemical tracer arrived slightly after the tritium, but this was presumably due to adsorption by the relatively large amounts of lignite present in the sand. In clean sand, the movement of the chemical tracer follows the movement of the water very closely (See Figure 22).

3.2.4 Introduction of the chemical barrier and performances

Before the introduction of the necessary reagents a tracer test was performed on the site where the chemical barrier would be installed.

As a result of this test it was concluded that a serious inhomogeneity, due to lignite, existed at that place as a double ^{3}T activity peak was detected in the pumped well water. The soil was afterwards washed with NaOH removing almost entirely the lignite present. A second tracer test after the cleaning procedure gave a regular result with a single ^{3}T peak.

The 3 wells for injection of chemicals are each 0.75 m apart and six meter below soil surface. Observation dry holes each 0.75 m apart are placed on two rows parallel to the injection wells at a distance of 0.75 m on both sides. The total width is 3 m, the depth 3 to 6 m and the thickness 1 m; making up an effective volume of about 9 m^3 in the aquifer or 4 m^3 void volume.

In order to be able to compare the results of the ^{85}Sr migration before and after the introduction of the barrier, a simultaneous injection of 50 mCi ^{85}Sr, 2 Ci ^{3}T and chemical tracer was carried out just before the injection of the chemicals.

The results of this test, shown on figure 23, indicate that the ^{85}Sr breaks through with the ^{3}T and the chemical tracer. The cleaned white sand does not show any retention capacity.

The first chemical to be injected in the aquifer was 5 m^3 10% HF prepared from 800 kg commercially available 70% HF. Using an acid resistant pump the solution was injected at a rate of 1 m^3/h and left for contact with the sand particles during 48 h.

Thereafter 5.6 m^3 of a 0.05 M $K_2H_2Sb_2O_7$ solution (about 1.5 void volume) was introduced according to the same method. This solution was tagged with ^{124}Sb in order to follow the distribution of the reagent in the aquifer. By measuring the activity in the dry holes it was found that the central part of the barrier was homogeneously filled with the $K_2H_2Sb_2O_7$ reagent.

About 24 hours after the injection of the antimony salt, a mixture of 2 Ci ^{85}Sr, 2 Ci ^{3}T and a quantity of chemical tracer was injected into a well 1.5 m from the chemical barrier. The tracercloud was pumped through the barrier at a rate of 1.25 m^3/h by a well situated also at 1.5 m from the axis. This speeding up of the migration rate was necessary for reasons of programme. The breakthrough curves of ^{3}T and chemical tracer are identical to those in figure 23. The ^{85}Sr breakthrough is plotted on figure 24.
The first traces of ^{85}Sr appear after 180 m^3 and a maximum was found after 600 m^3.

The retention properties of the barrier is approximately 20 times higher than the natural environment. This figure being about 50% lower than the laboratory result proves yet that the principle of the chemical barrier is applicable to sandy aquifers. By investigating the different hydrologic parameters as pumping rate, dilution of chemicals and reaction period it must be possible to approach the laboratory results.

The delay imposed upon the ^{90}Sr migration by the chemical barrier has two important advantages :

1. the intervention crew is able to prepare thoroughly the final decontamination procedure,
2. the contaminated zone which was too large to treat by conventional pumping is concentrated in a relatively small volume which is easy to decontaminate.

A barrier of 2 m thickness can concentrate a contaminated zone smeared out over a distance of 40 to 100 m according as the results of the field or laboratory tests are taken into account. And consequently the delay for intervention is multiplied by the same factors (20 to 50).

If a barrier of 50 m width on 2 m thickness should be installed up to a depth of 10 m it would be necessary to treat a volume of 1000 m^3 corresponding to a void of 400 m^3. A factor of 1.5 has to be applied in order to insure proper saturation of the zone.

4. CONCLUSIONS

4.1 Hydrologic investigations

The experimental drilling on the CEN site has confirmed the statement of Gulinck et al that the groundwater aquifer is made up of two sand formations, the Mol sand up to 9 meters and the Kasterlee sand between 9 and 25 meters [3]

The discussions about the permeability of the contact layer between the groundwater aquifer and the underlying have not been classified by the recent drillings. The most probable conclusion in accordance with most observations is the existence of a more or less impermeable boundary between Kasterlee and Diestian made up of tiny layers of micaceous material and clay. The vertical discharge through this contact zone could not be determined.

The permeability coefficient in the Mol sand amounts to 1.16 cm/min which is in agreement with the previous data. The Kasterlee sand is about two times less permeable and the permeability coefficient is 0.56 cm/min.

4.2 Scavenging well tests

Using the information obtained during the hydrogeologic investigations a series of pumping tests were designed in order to test the feasibility of removing the radioactive contamination from the water-table. The results of four tests in the upper and lower part of the aquifer show that the total amount of water to be pumped in order to remove a contaminated zone containing water tracers is made up of two contributions which are roughly 50-50% for the Mol sand and Kasterlee sand. This ratio is determined by the depth of the scavenging well, the thickness of the two formations crossed and the permeabilities.

Based on the results obtained some design calculations are presented for the installation of scavenging well systems in the water-table aquifer. Where a relatively large area (300 m x 300 m) would be contaminated, years of pumping are necessary in order to remove the contamination.

4.3 Chemical barriers

Laboratory tests have shown that by hydrolysis of $K_2H_2Sb_2O_7$ in acid medium a coating can be formed around the sand particles which has specific ion exchange properties of Sr. About 60 pore volumes of water tagged with ^{85}Sr could be percolated through a unit volume of pretreated sand before breakthrough of the radionuclides occurs.

Field tests on the penetration of reagents into the aquifer have indicated that 1.5 to 2 void volumes have to be injected in order to get an almost complete saturation of the medium.Zones with different permeabilities as is the case at Mol where two sand layers occur have to be treated separately in order to keep the reagent consumption at its minimum. A detailed knowledge of the geologic structure of the strata is required if a valuable design has to be made. Successful tests have been performed on the removal of lignite layers with caustic soda; this pretreatment was necessary in order to have a "blank" medium without any adsorption capacity for Sr. Injections of water tracers and ^{85}Sr in the medium where the barrier was to be installed have proven that the sand did not adsorb any radionuclide.

The barrier was installed into the aquifer at a depth of 6 m by injecting first concentrated HF (40%) followed by 0.05 M $K_2H_2Sb_2O_7$. The barrier was homogeneous as could be deduced from the distribution of ^{125}Sb injected with the reagent. About 2 Ci of ^{85}Sr were injected at one side of the barrier and pumped through; the peak of activity appeared 20 times later than in the similar test before the barrier was installed.

Economical computations about the design of large scale barriers have led to the conclusion that the installation of such barrier only can be conceived when relatively small volumes have to be treated e.g. around storage tanks.

Both remedial techniques : the scavenging and barrier building, may be considered as complementary techniques and not as exclusive.

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TABLE I

Summary of Results of Pumping Tests, May 16, 1966

	Transmissibility (m^2/hour)	Permeability (cm/min)	Storage Coefficient
Mol Sand			
Well 5S	4.10	1.14	0.059
Well 10S	4.01	1.115	0.0631
Well 15S	4.38	1.22	0.614
Average	4.16	1.16	0.0612
Kasterlee Sand			
Well 5D	6.12	0.537	0.0088
Well 10D	6.30	0.552	0.0148
Well 15D	6.80	0.596	0.0156
Average	6.41	0.562	0.0131

TABLE II

Laboratory investigation on the efficiency of various salts for building chemical barriers into porous media.

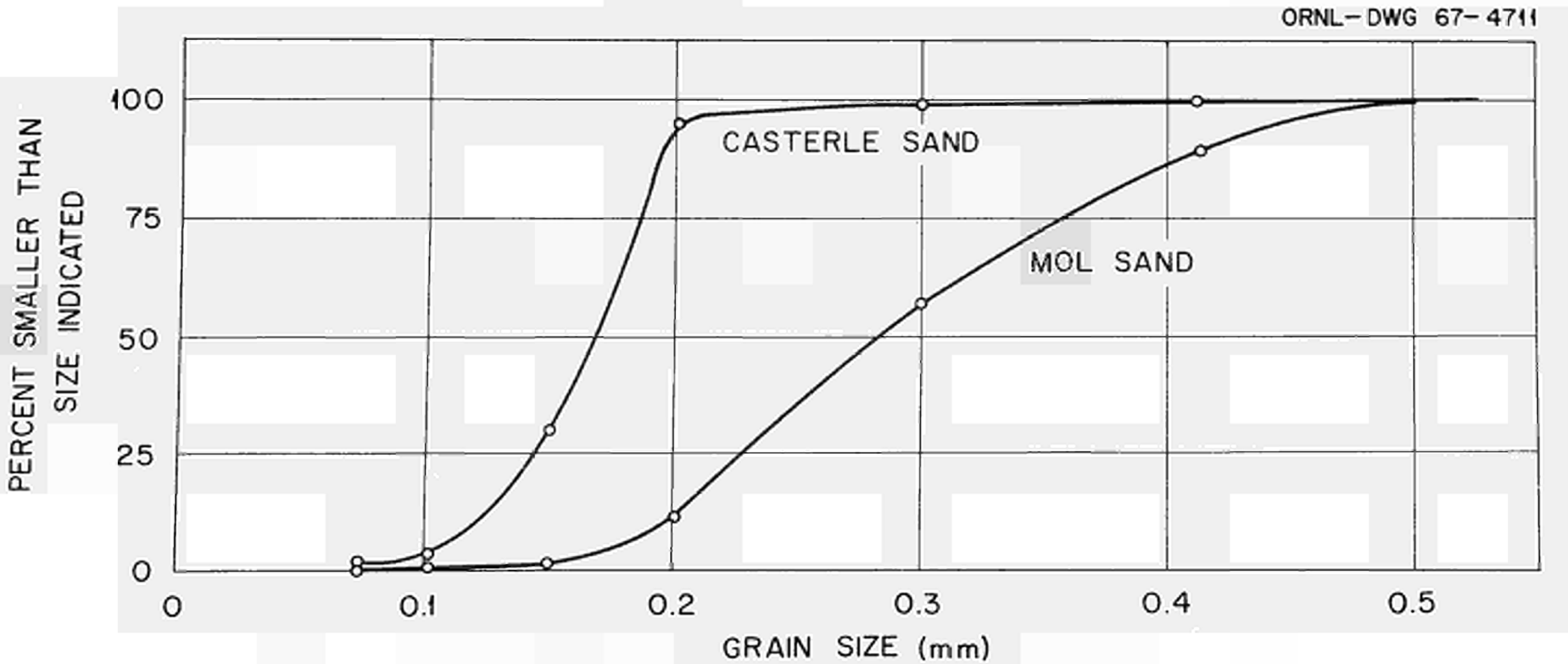
Concentration of etching reagent (2 void volumes added)	Hydrolyzable reagent	Concentration M	Reaction period (hours)	Number of void volumes tagged with Sr adsorbed by the treated soil
30% HF	$FeCl_3$ + NaOH	1	12	15
30% HF	$NaSnO_3$ $3H_2O$	1	12	9
30% HF	$NaSnO_3$	1	15	37
30% HF	$K_2H_2Sb_2O_7$	0.05	18	63
30% HF	$K_2H_2Sb_2O_7$	0.025	8	43
20% HF	$K_2H_2Sb_2O_7$	0.05	14	50
10% HF	$K_2H_2Sb_2O_7$	0.1	24	64
5% HF	$K_2H_2Sb_2O_7$	0.05	24	56
2% HF	$K_2H_2Sb_2O_7$	0.1	24	28
Cp4 1/4	$K_2H_2Sb_2O_7$	0.1	24	43
Cp4 1/2	$SnCl_2$ + NH_4OH	0.5	24	27

TABLE III

Uptake capacity of a polyantimonic barrier for Sr and its influence on the permeability

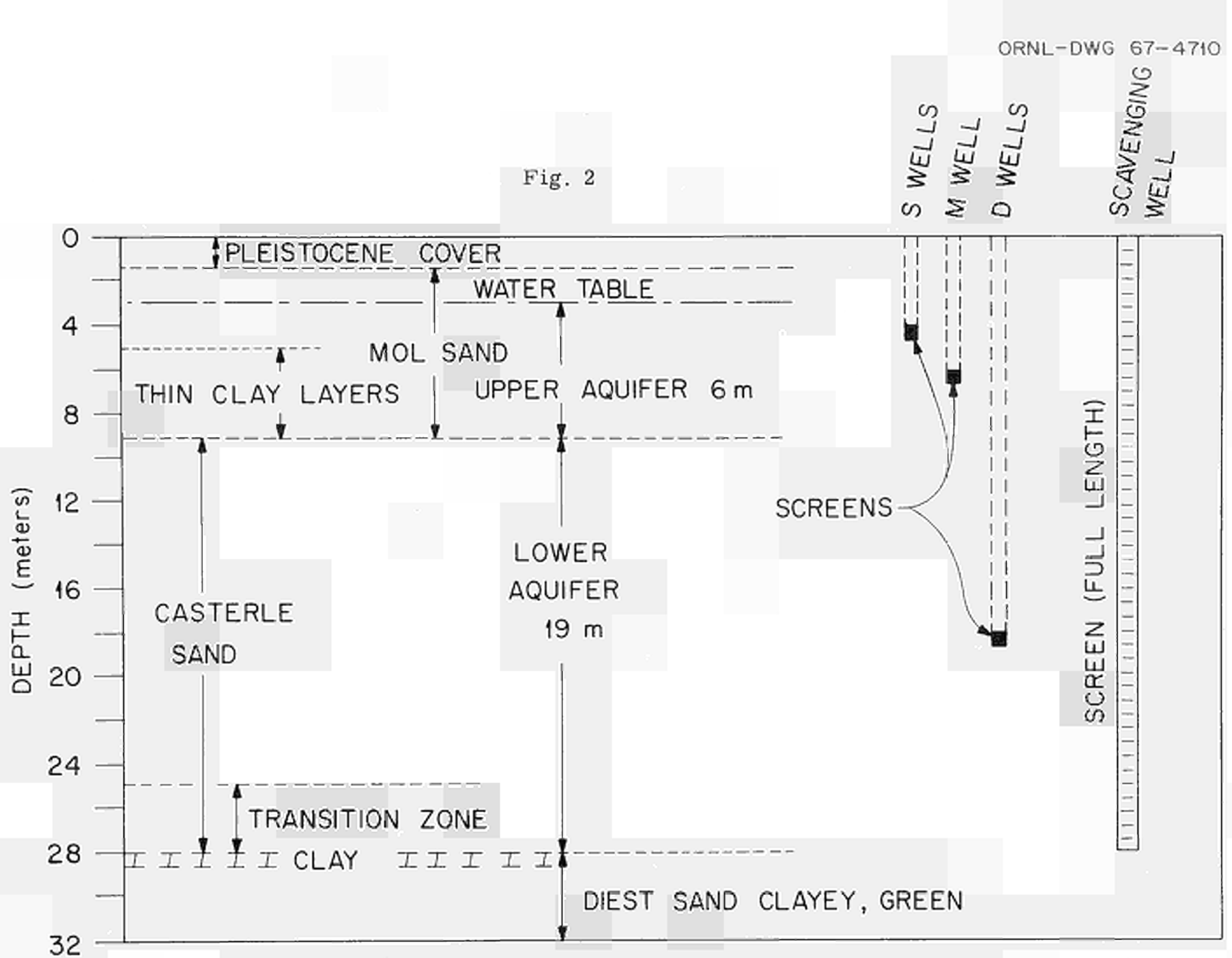
Concentration of HF %	$K_2H_2Sb_2O_7$ Moles/l	Uptake capacity bedvolumes before breakthrough	Influence on permeability
30	0.1	-	column clogged
25	0.1	28	decreased permeability
30	0.05	63	slightly decreased permeability
30	0.025	43	no change in permeability
20	0.1	59	decreased permeability
20	0.05	50	slightly decreased permeability
20	0.025	28	no change in permeability
10	0.1	64	slightly decreased permeability
10	0.05	53	no change in permeability
10	0.025	15	no change in permeability
5	0.1	49	no change in permeability
5	0.05	45	no change in permeability

Fig. 1



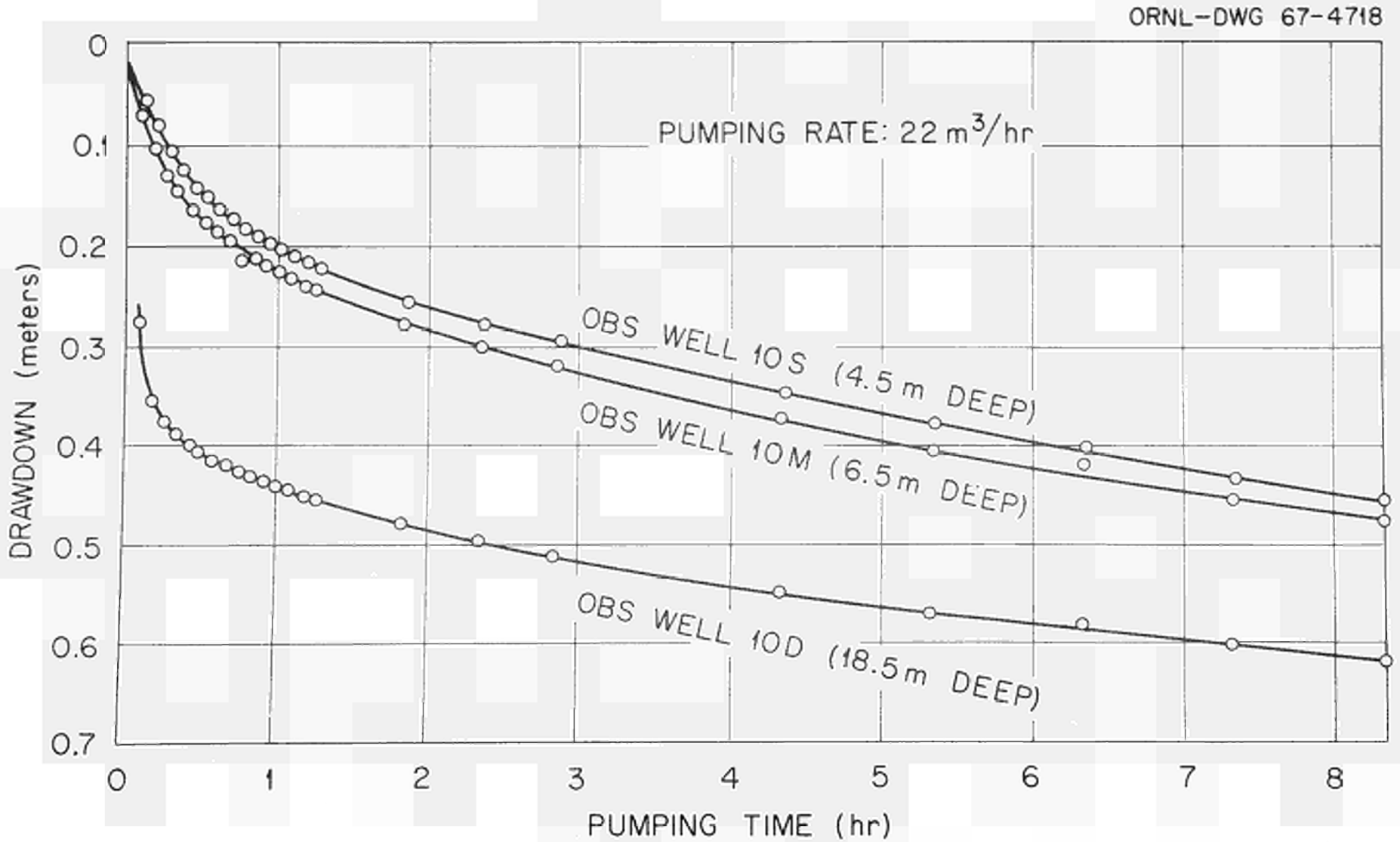
Comparison of Grain Size Distributions; Mol and Casterle Sands.

Fig. 2



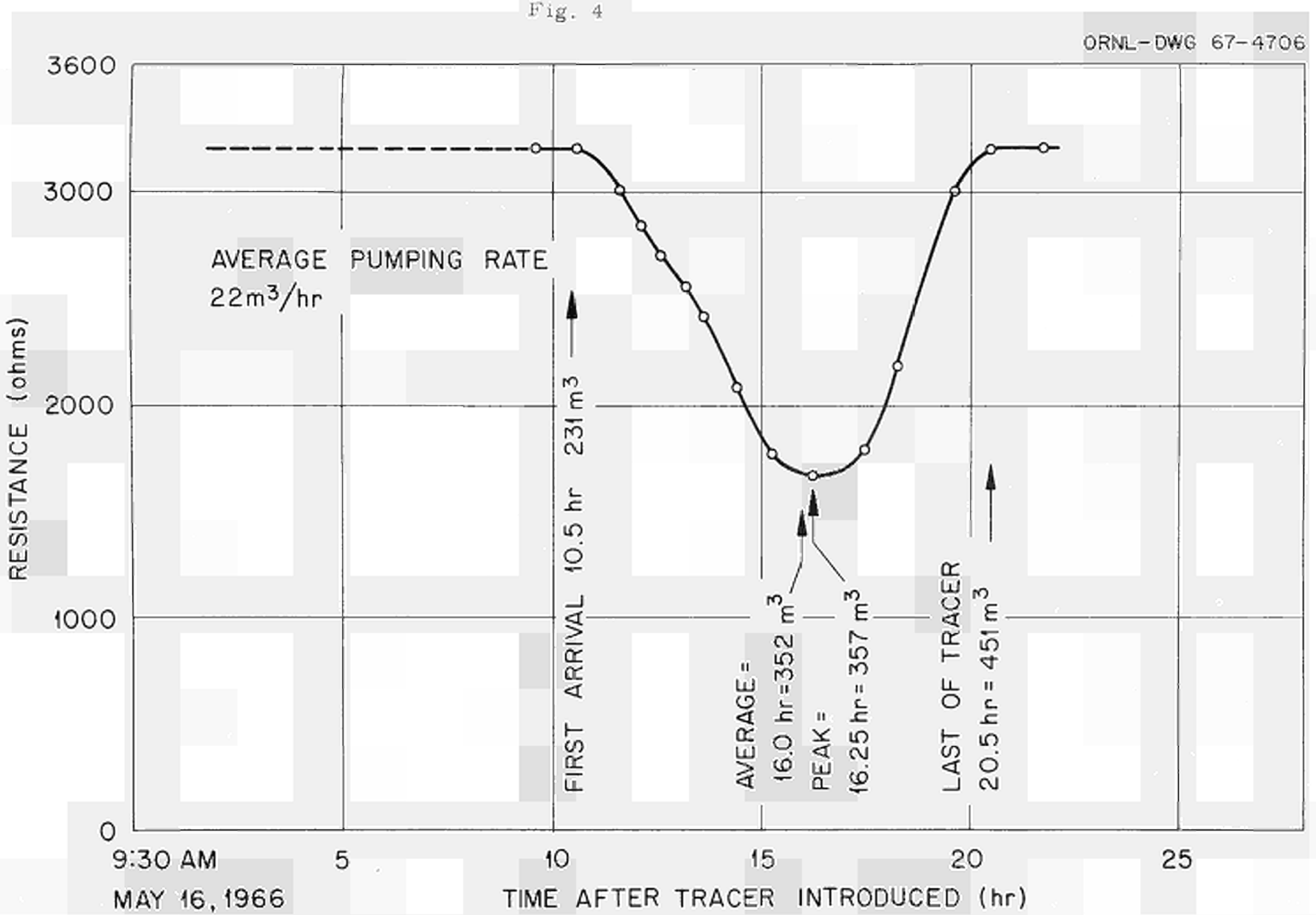
Stratigraphy and Well Depths Scavenging Test Site.

Fig. 3



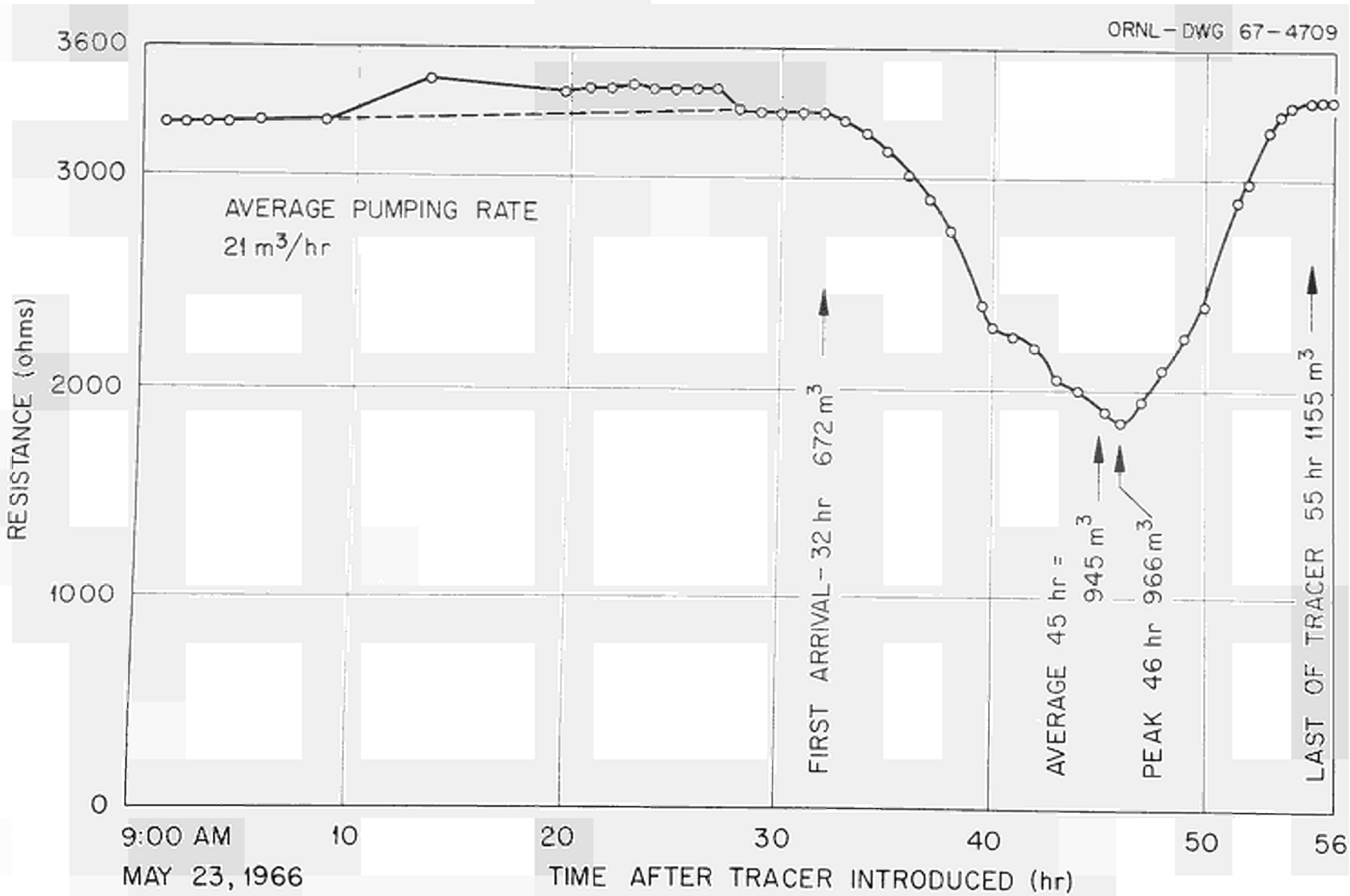
Comparison of Drawdowns in Observation Wells 4.5, 6.5 and 18.5 m Deep, Located 10 m From Scavenging Well, Aug. 17, 1966.

Fig. 4



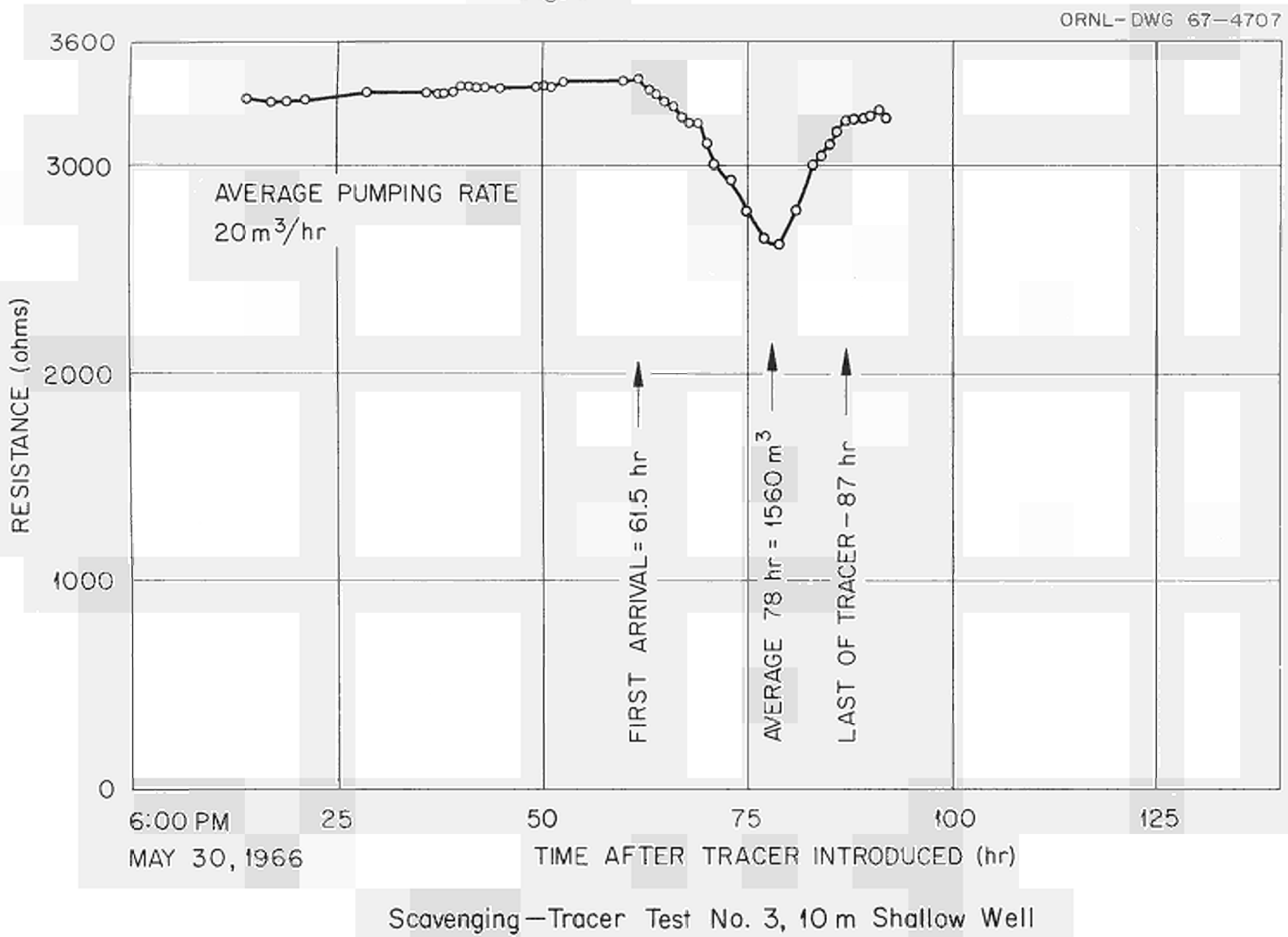
Scavenging—Tracer Test No. 1, 5 m Shallow Well.

Fig. 5



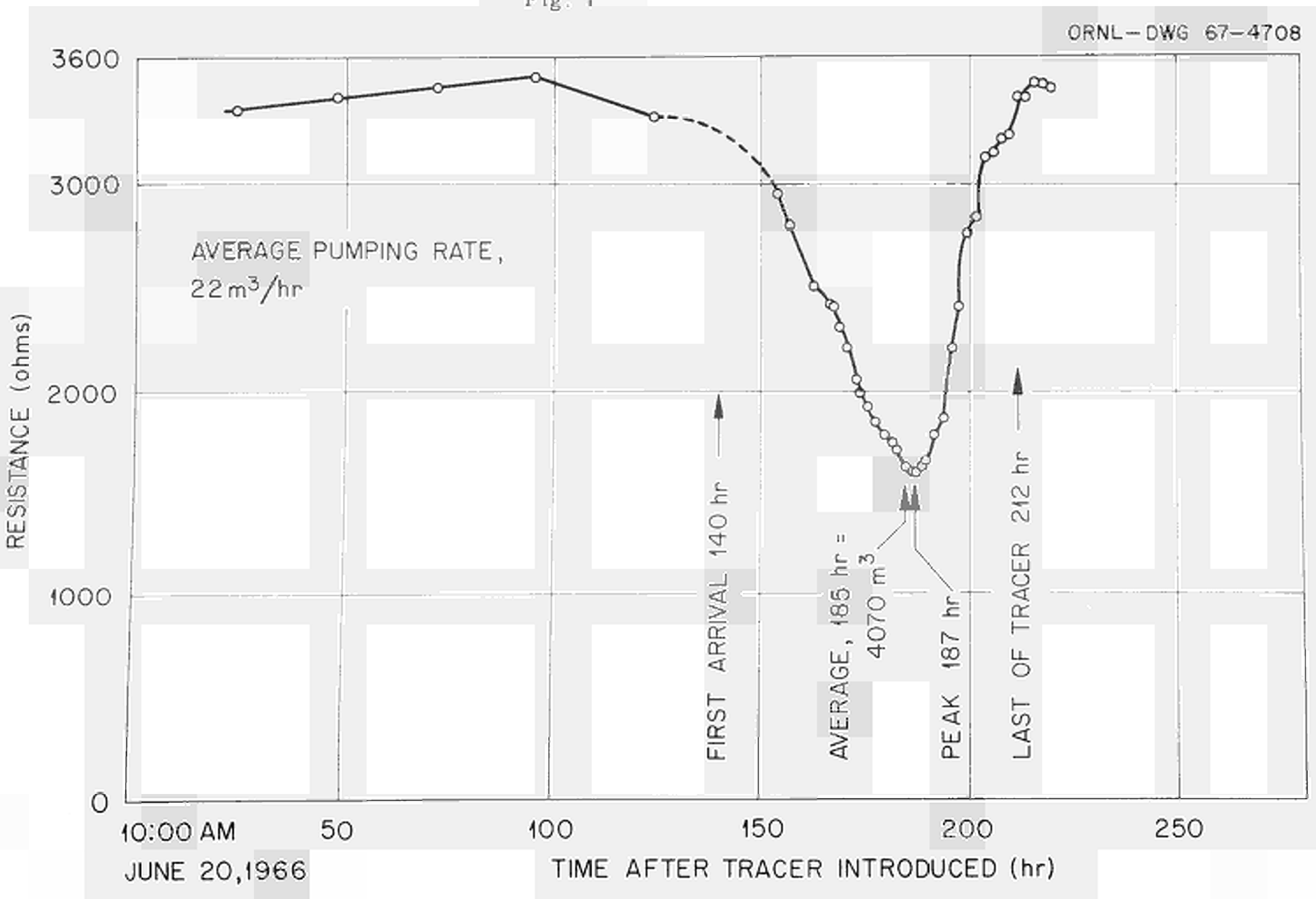
Scavenging-Tracer Test No. 2, 5m Deep Well.

Fig. 6



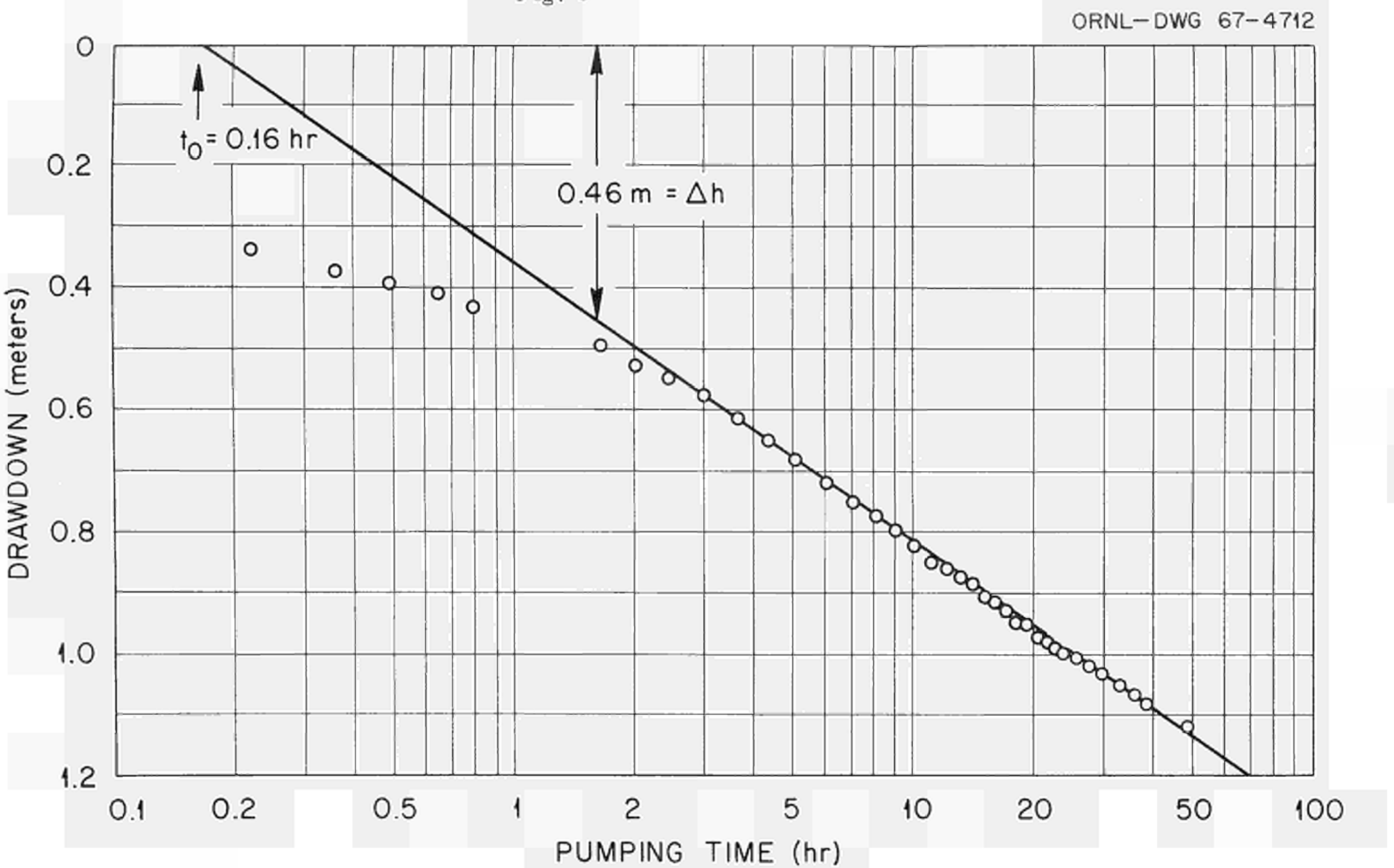
Scavenging—Tracer Test No. 3, 10 m Shallow Well

Fig. 7



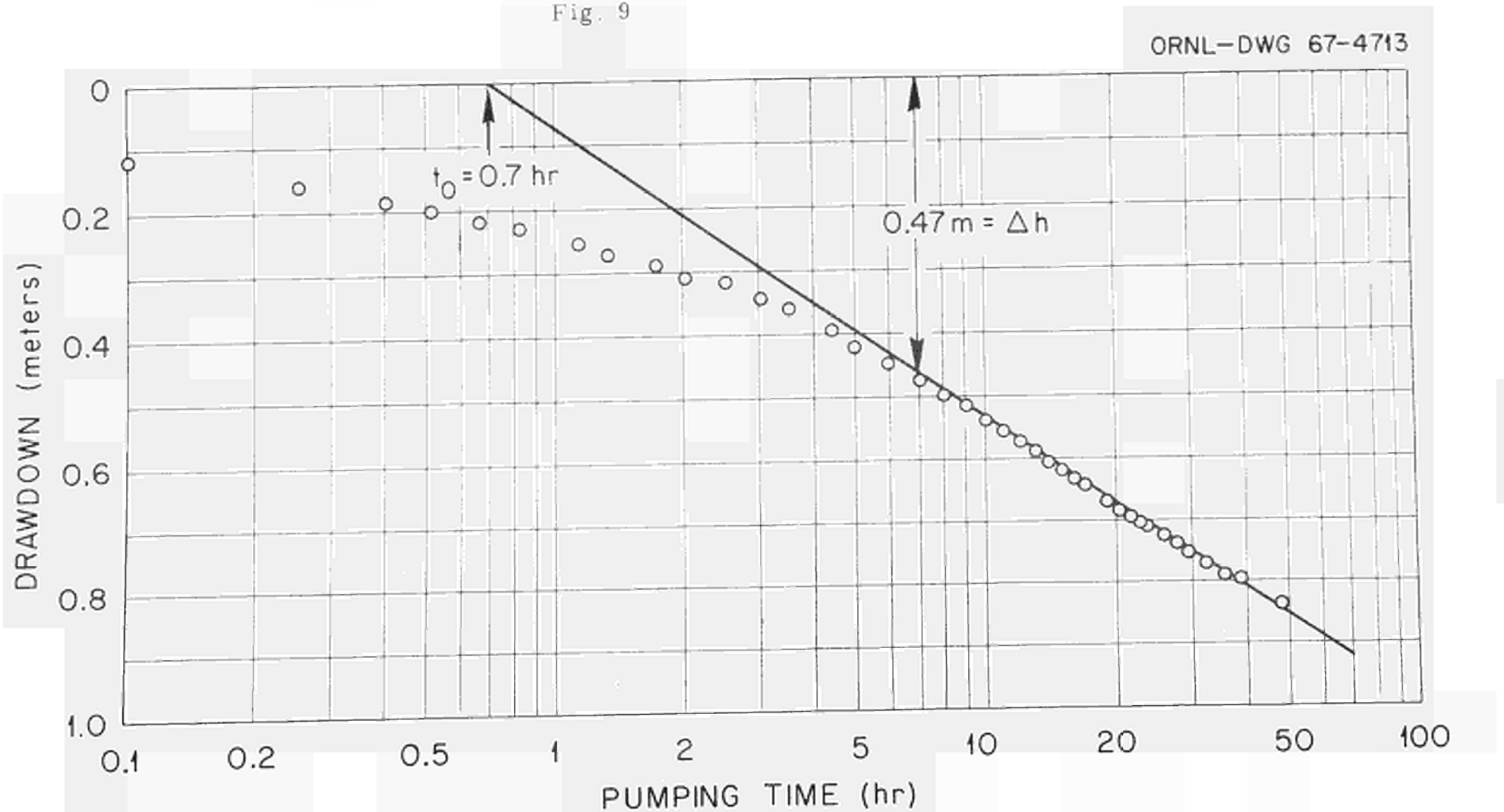
Scavenging—Tracer Test No. 4, 10 m Deep Well.

Fig. 8



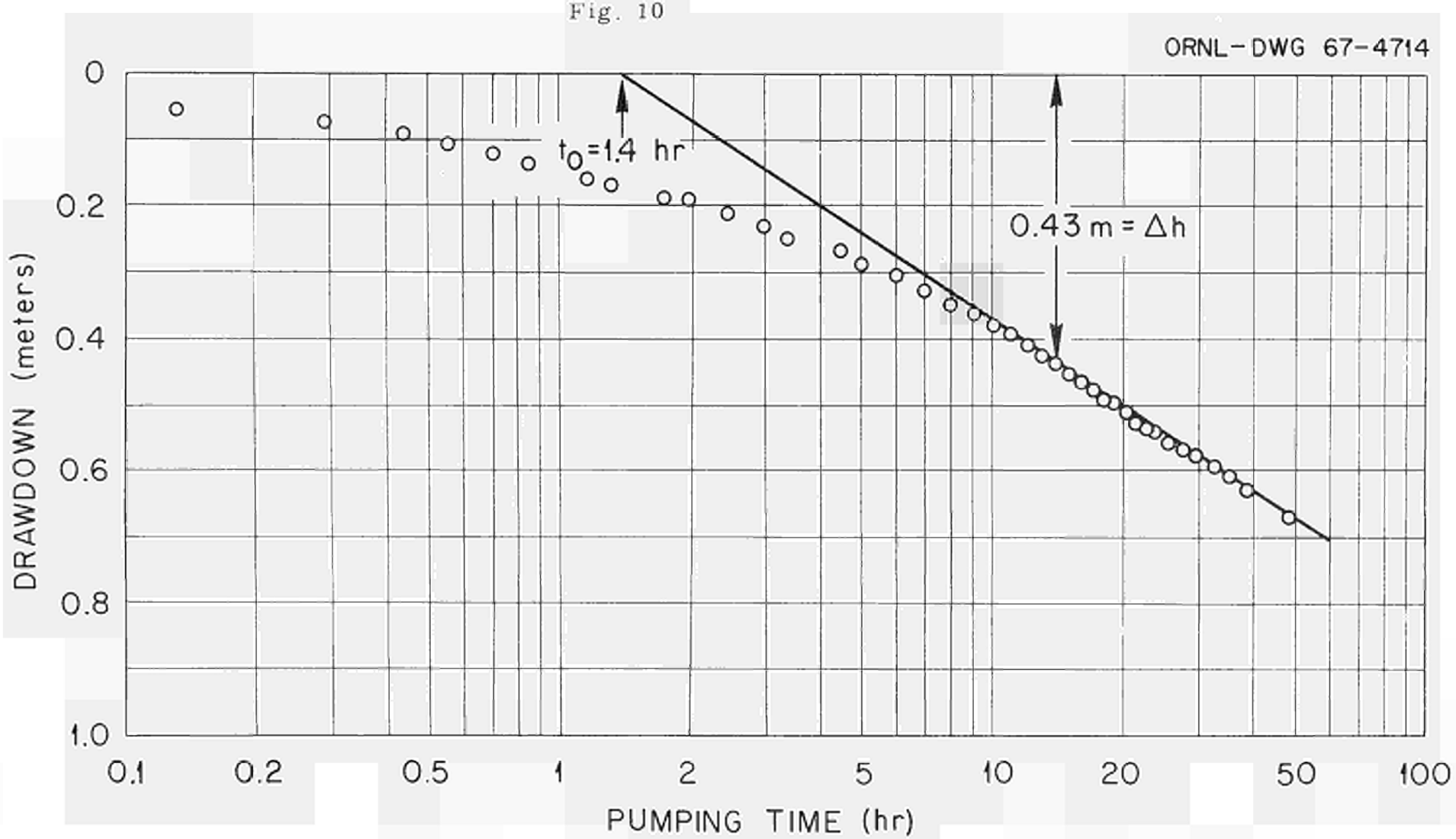
Pumping Test May 16, 1966; 5-meter Shallow Well.

Fig. 9



Pumping Test May 16, 1966; 10-meter Shallow Well.

Fig. 10



Pumping Test May 16, 1966; 15-meter Shallow Well.

Fig. 11

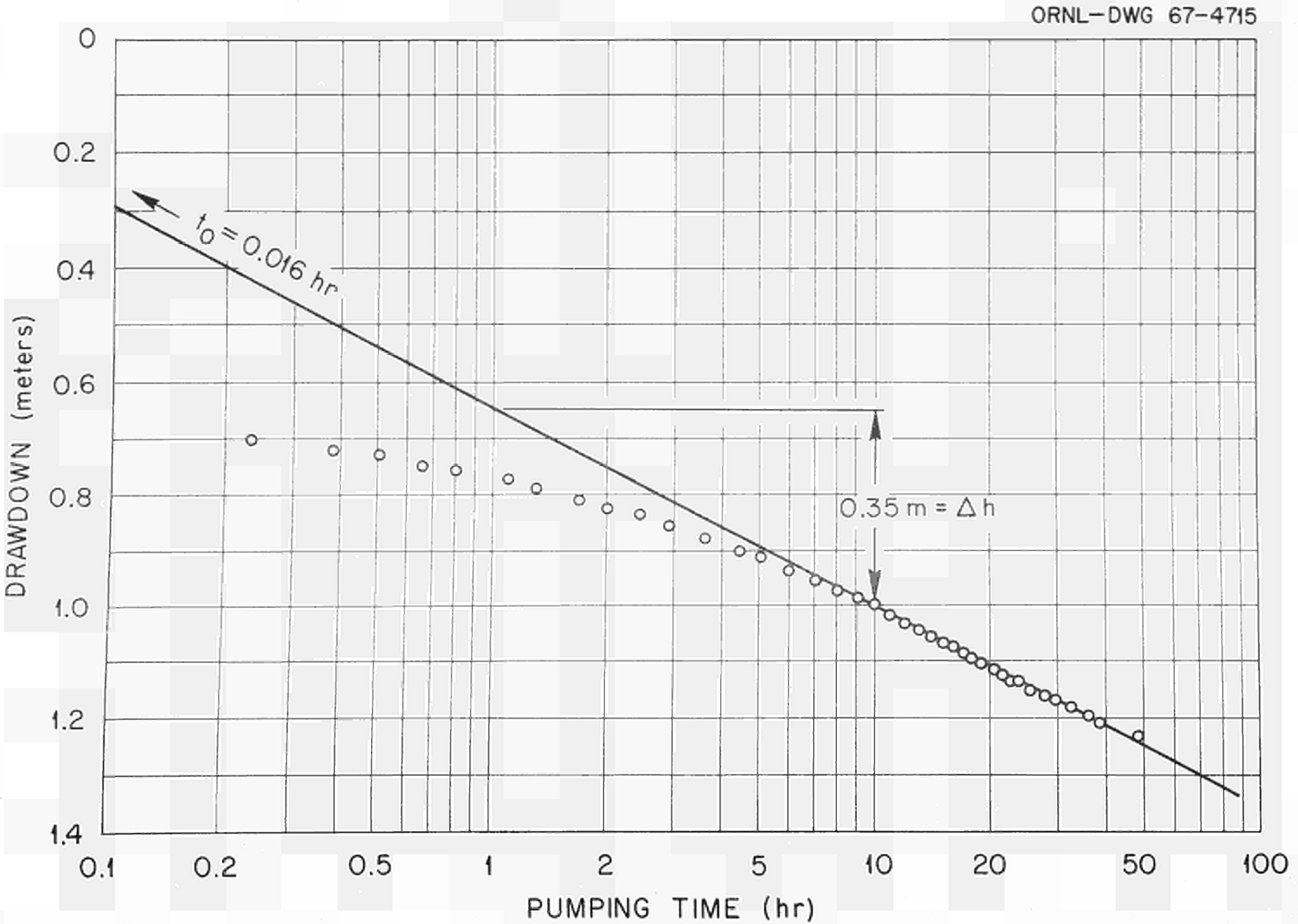
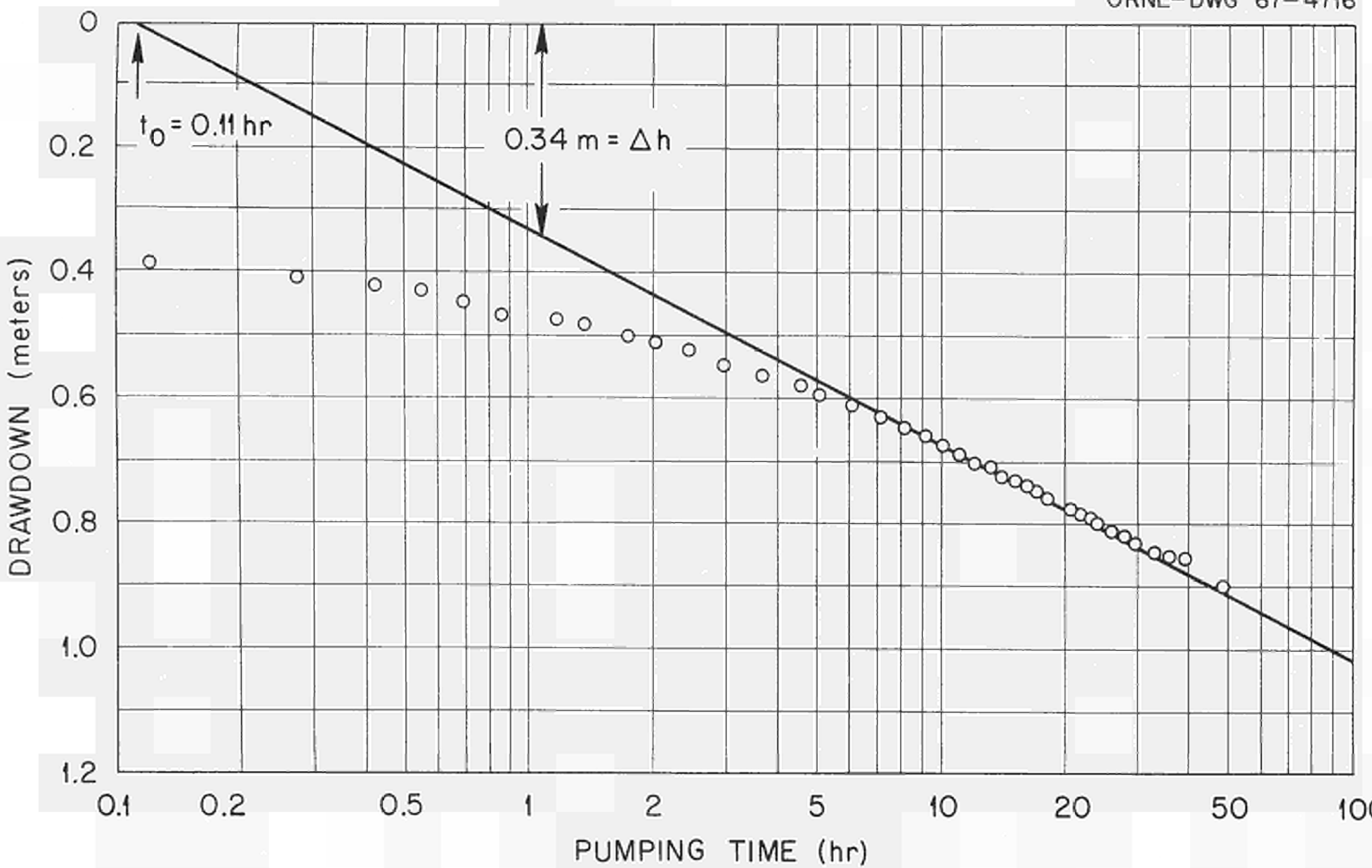
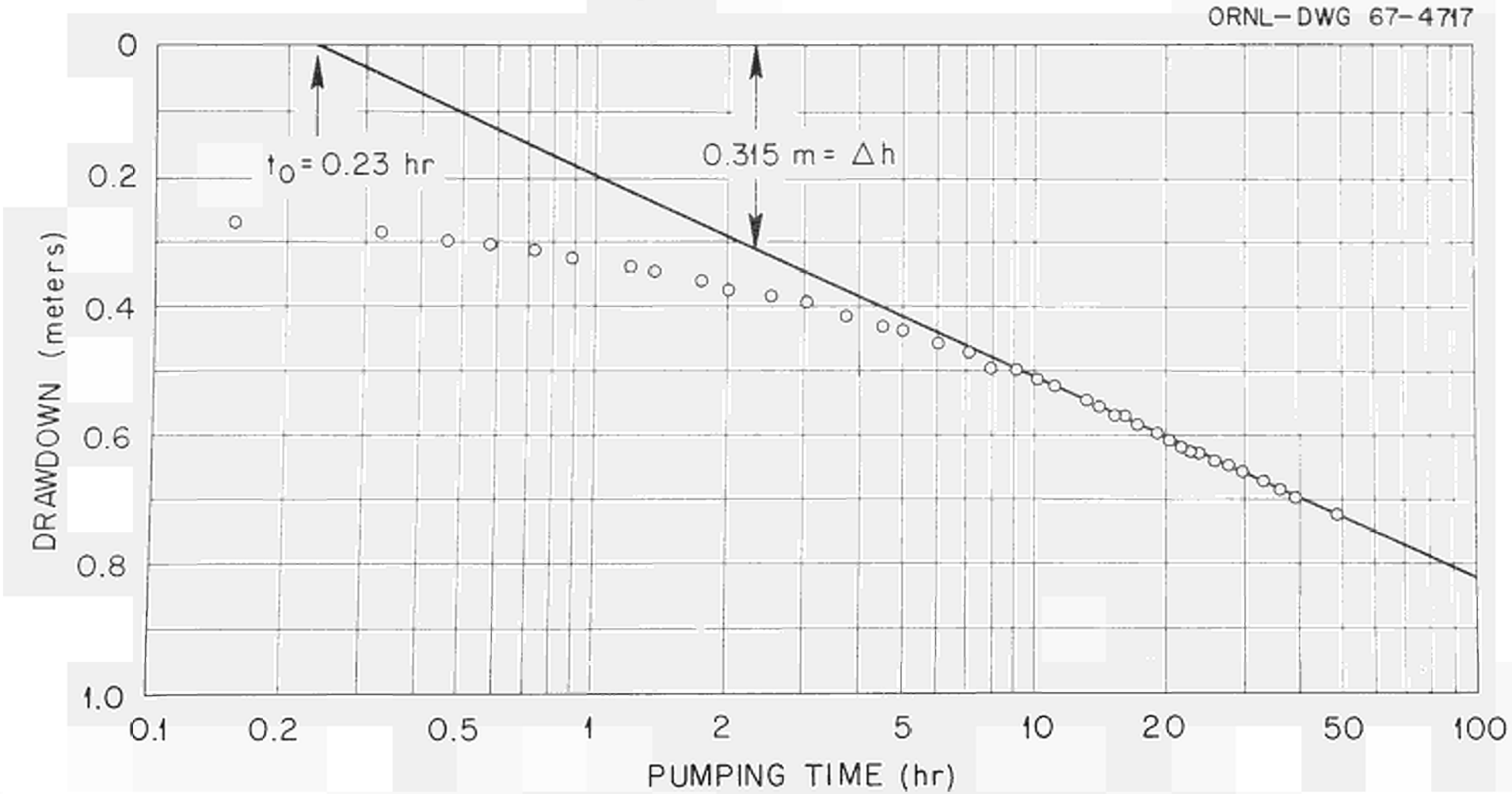

ORNL-DWG 67-4715
DRAWDOWN (meters)
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
t_o = 0.016 hr
0.35 m = Δh
0.1
0.2
0.5
1
2
5
10
20
50
100
PUMPING TIME (hr)

Fig. 12



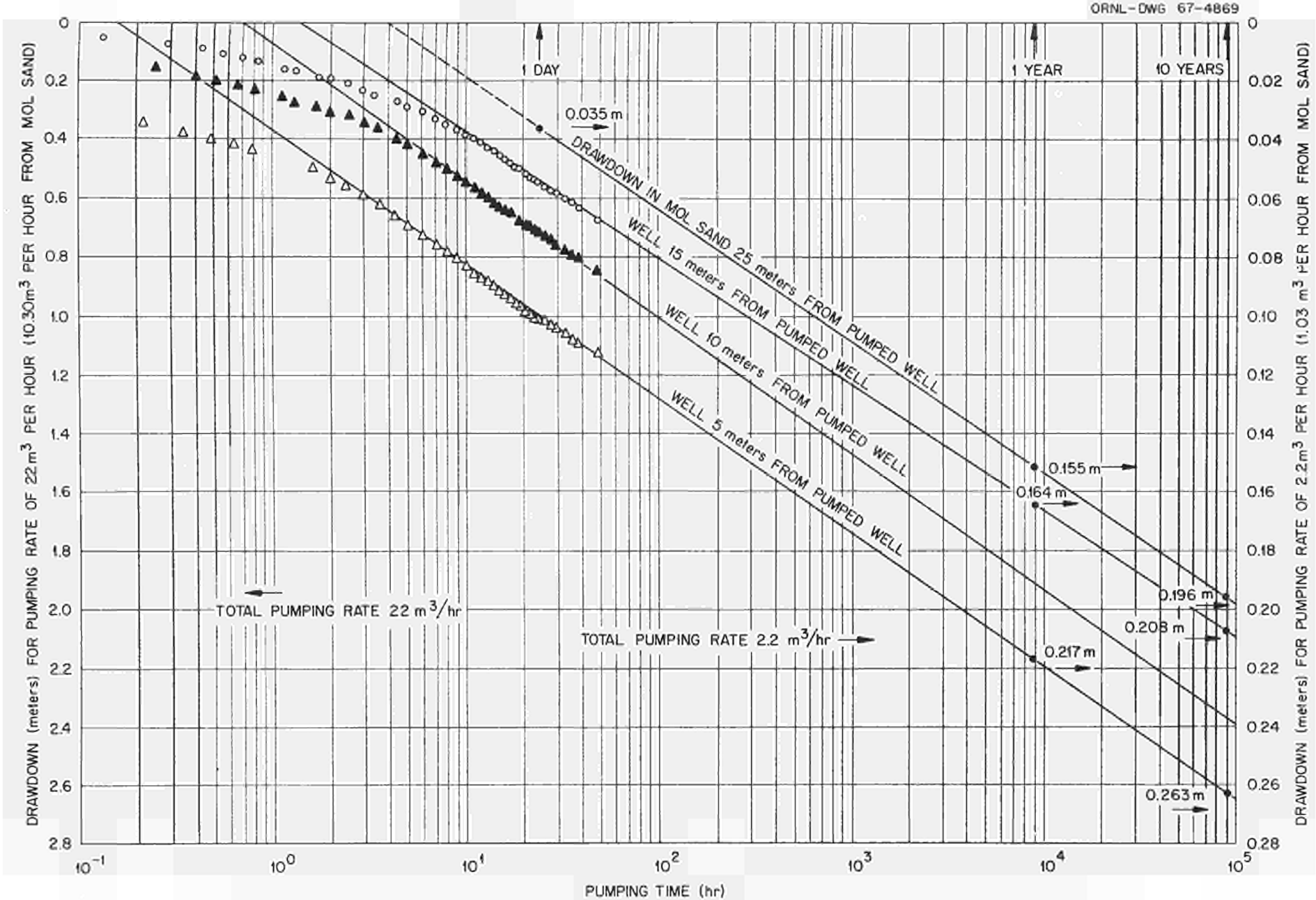
Pumping Test May 16, 1966; 10-meter Deep Well.

Fig. 13



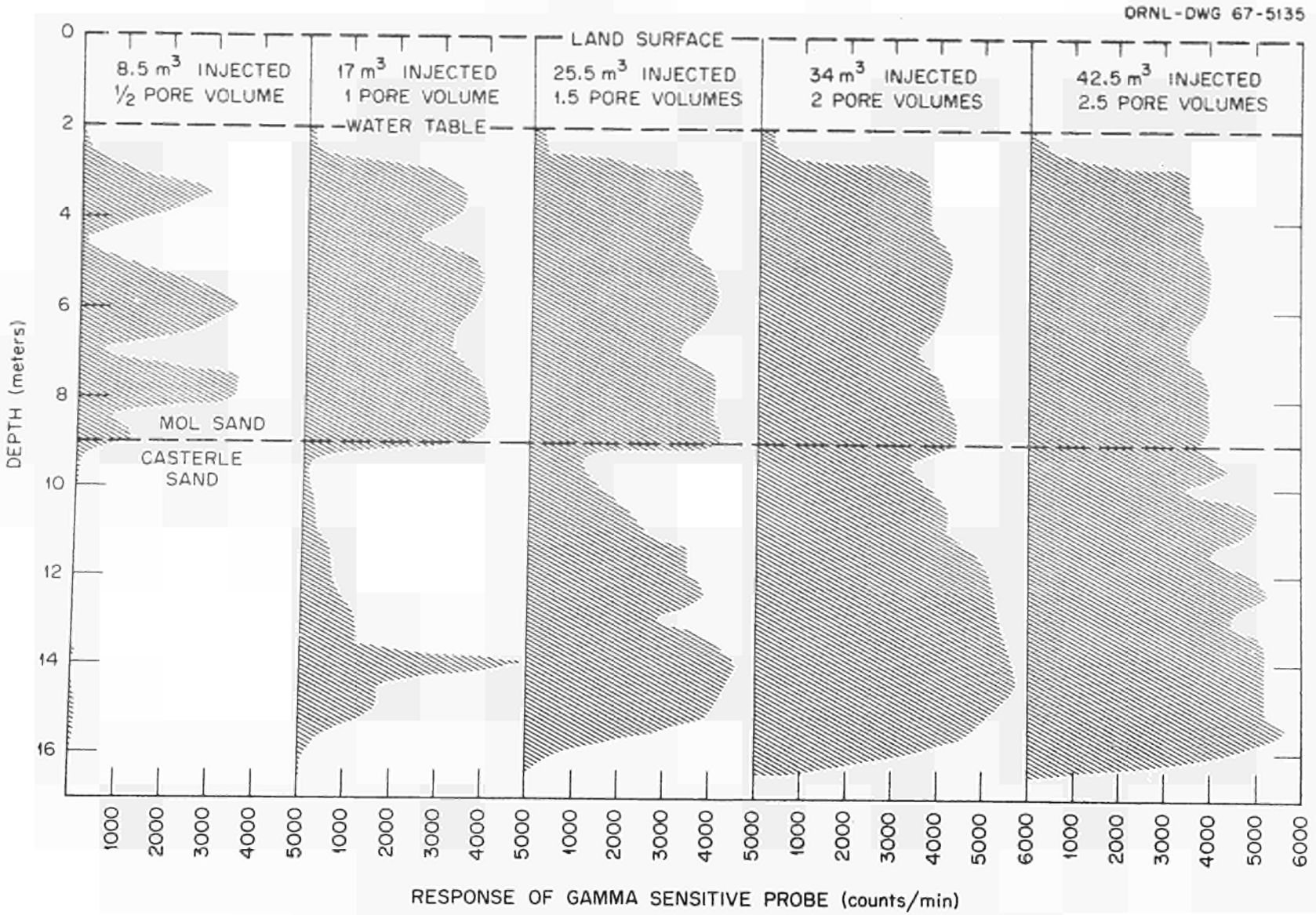
Pumping Test May 16, 1966; 15-meter Deep Well.

Fig. 14



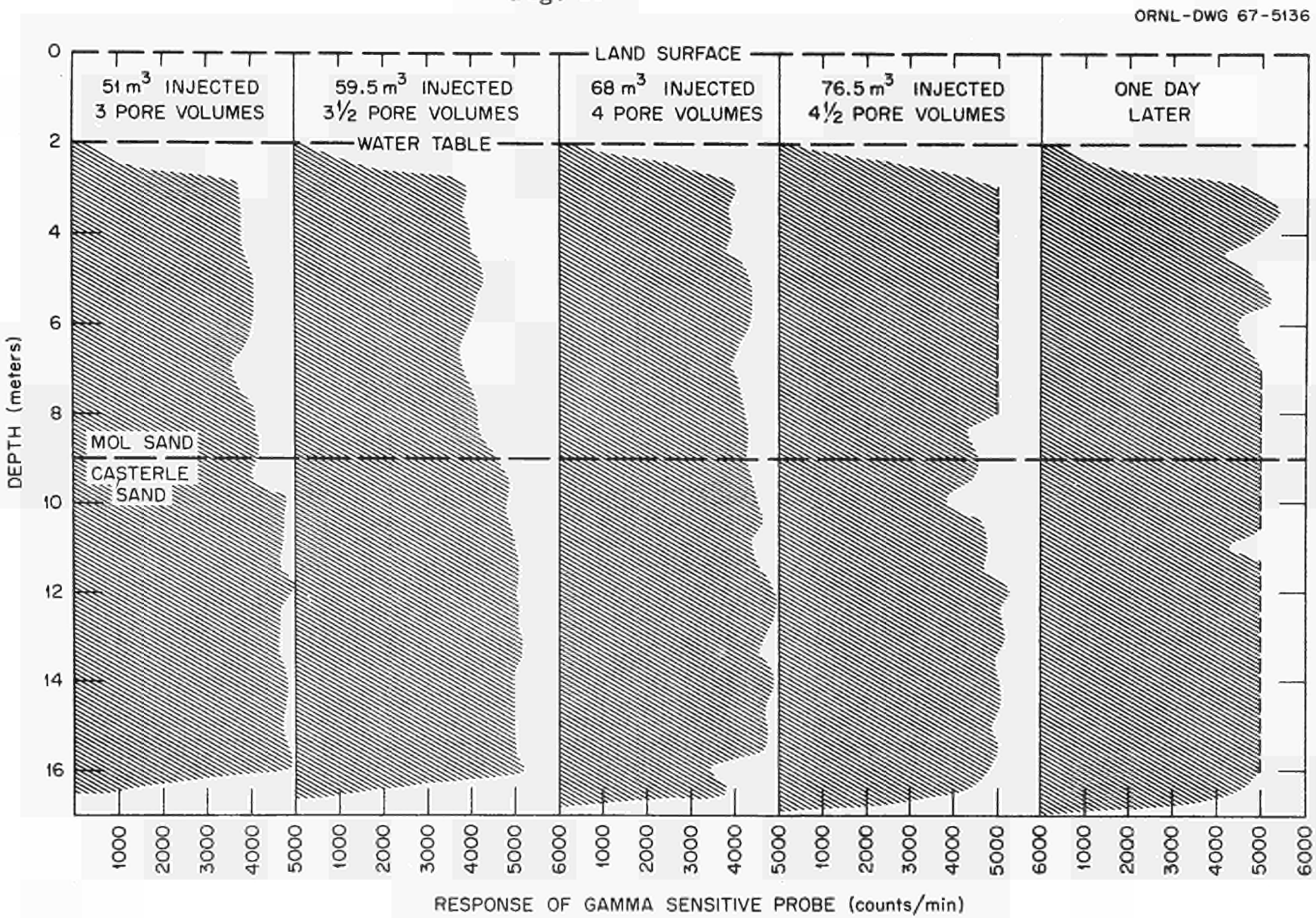
Lowering of Water Table in Upper (Mol Sand) Aquifer at Indicated Distances from Pumped Well.

Fig. 15



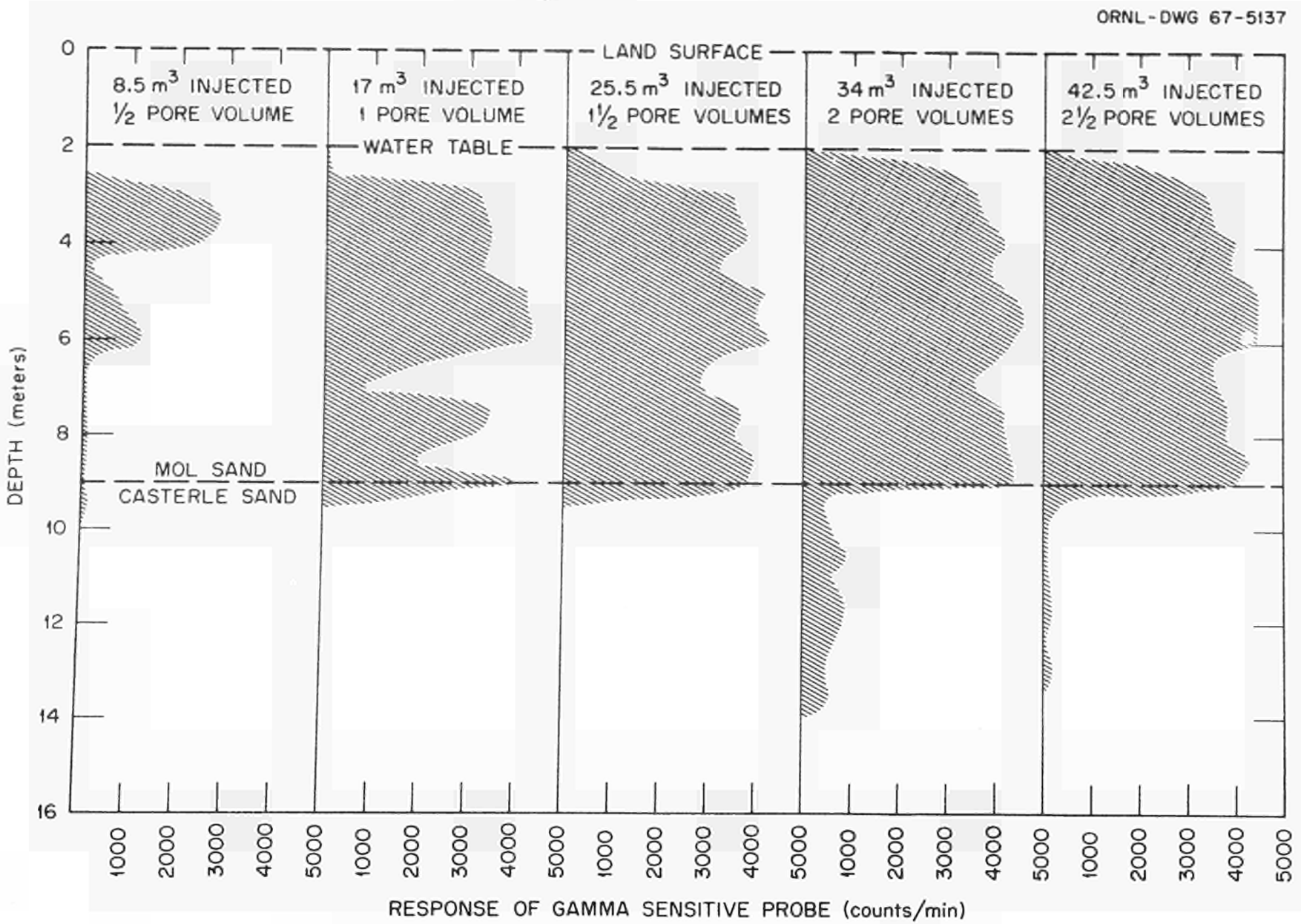
Tracer Injection Test; Dry Well 1 m West of Injection Well.

Fig. 16



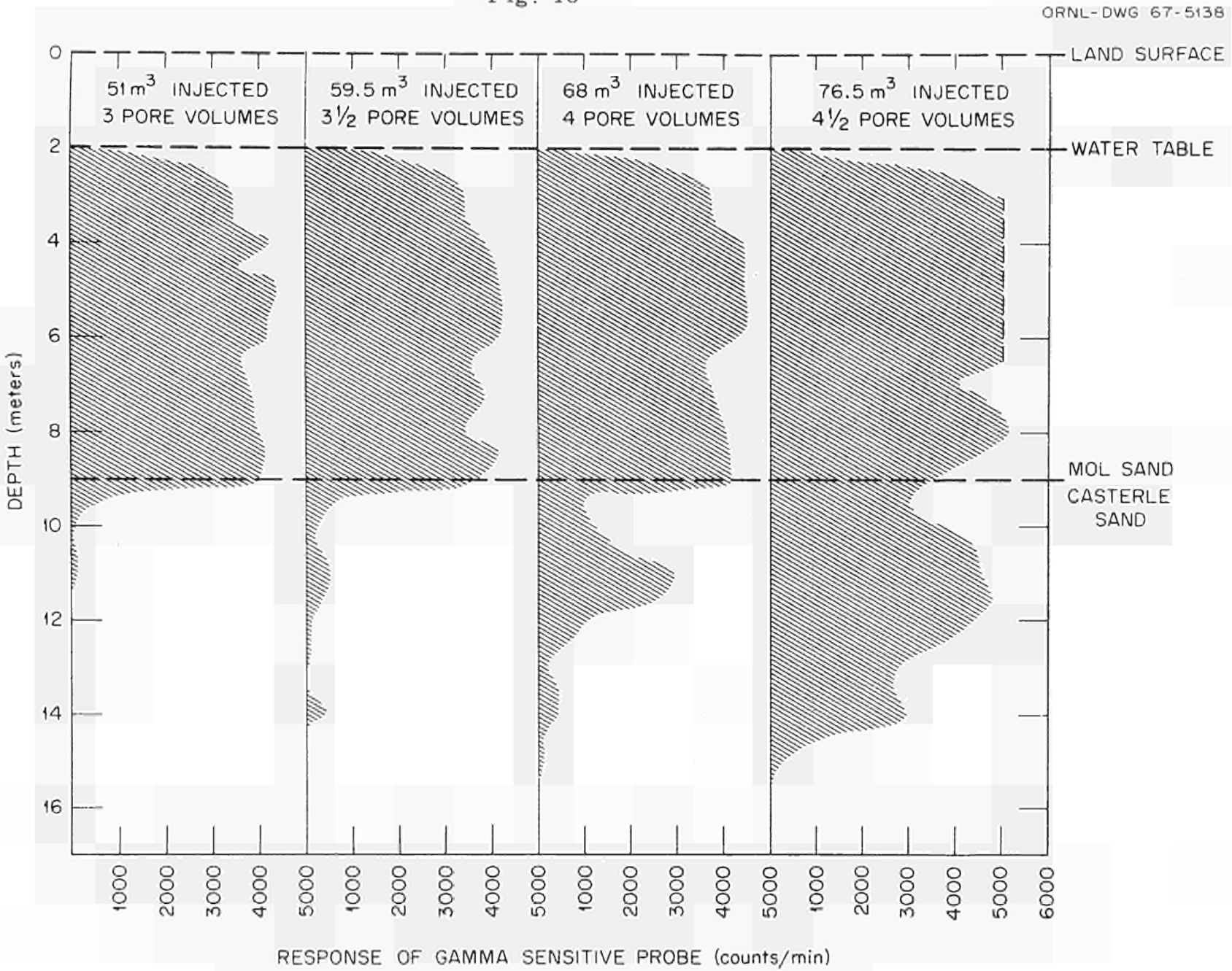
Tracer Injection Test; Dry Well 1m West of Injection Well.

Fig. 17



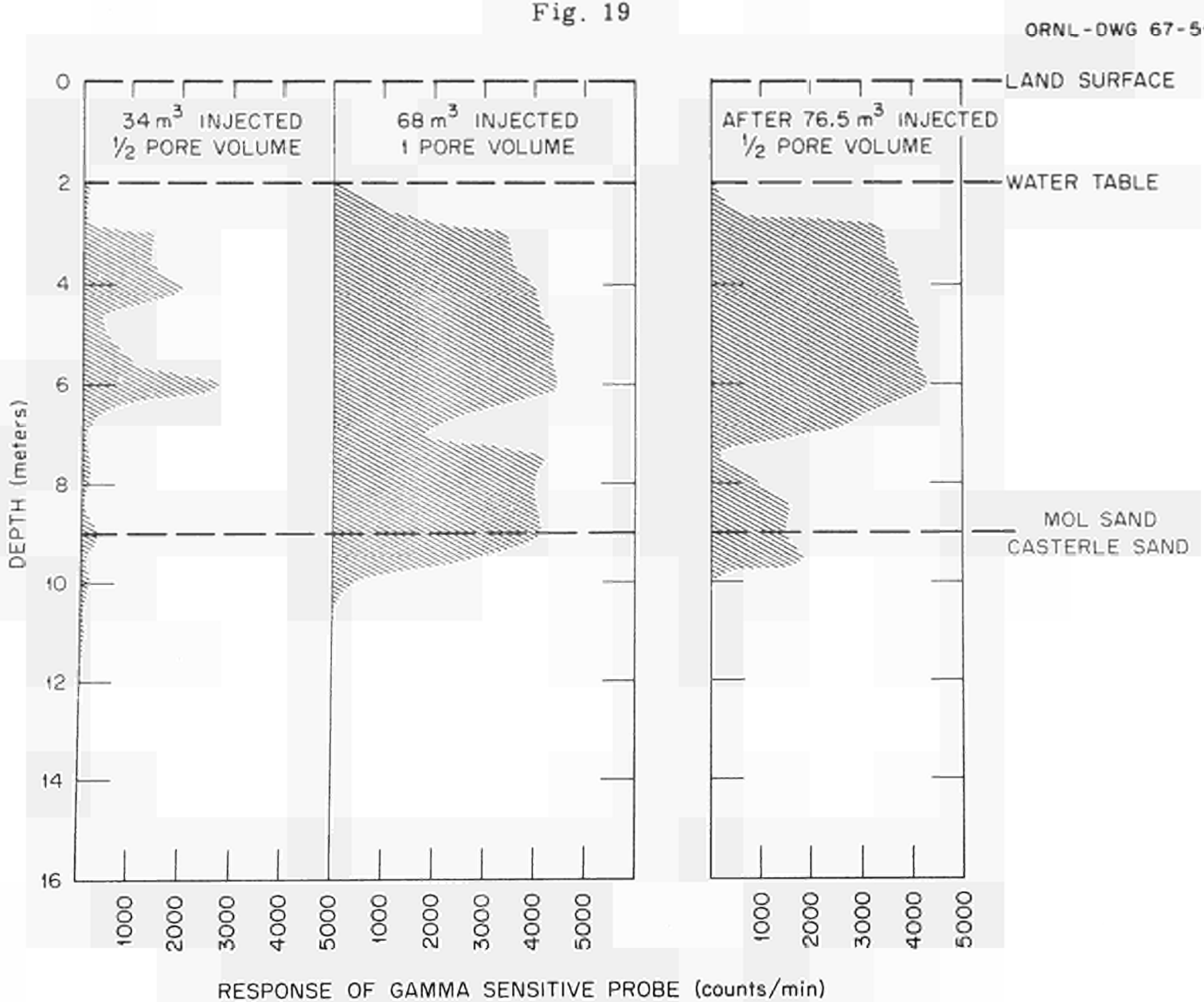
Tracer Injection Test; Dry Well 1m East of Injection Well.

Fig. 18



Tracer Injection Test; Dry Well 1m East of Injection Well.

Fig. 19



Tracer Injection Tests; Dry Well 2 m East of Injection Well.

Tracer Injection Tests; Dry Well 3 m East of Injection Well.

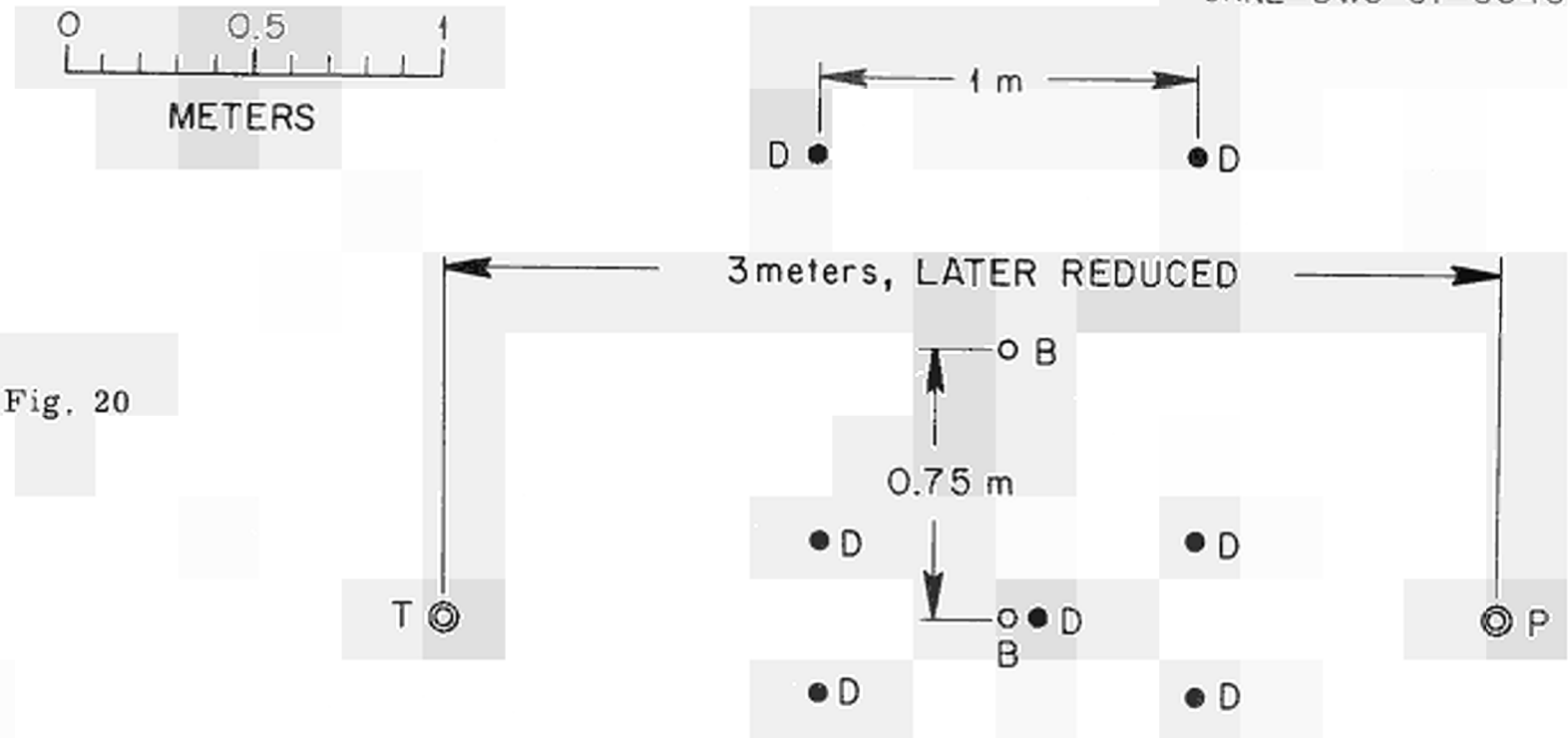


Fig. 20

◎ T TRACER-ISOTOPE INJECTION WELL
SCREENED 4.5 TO 5.0 meters

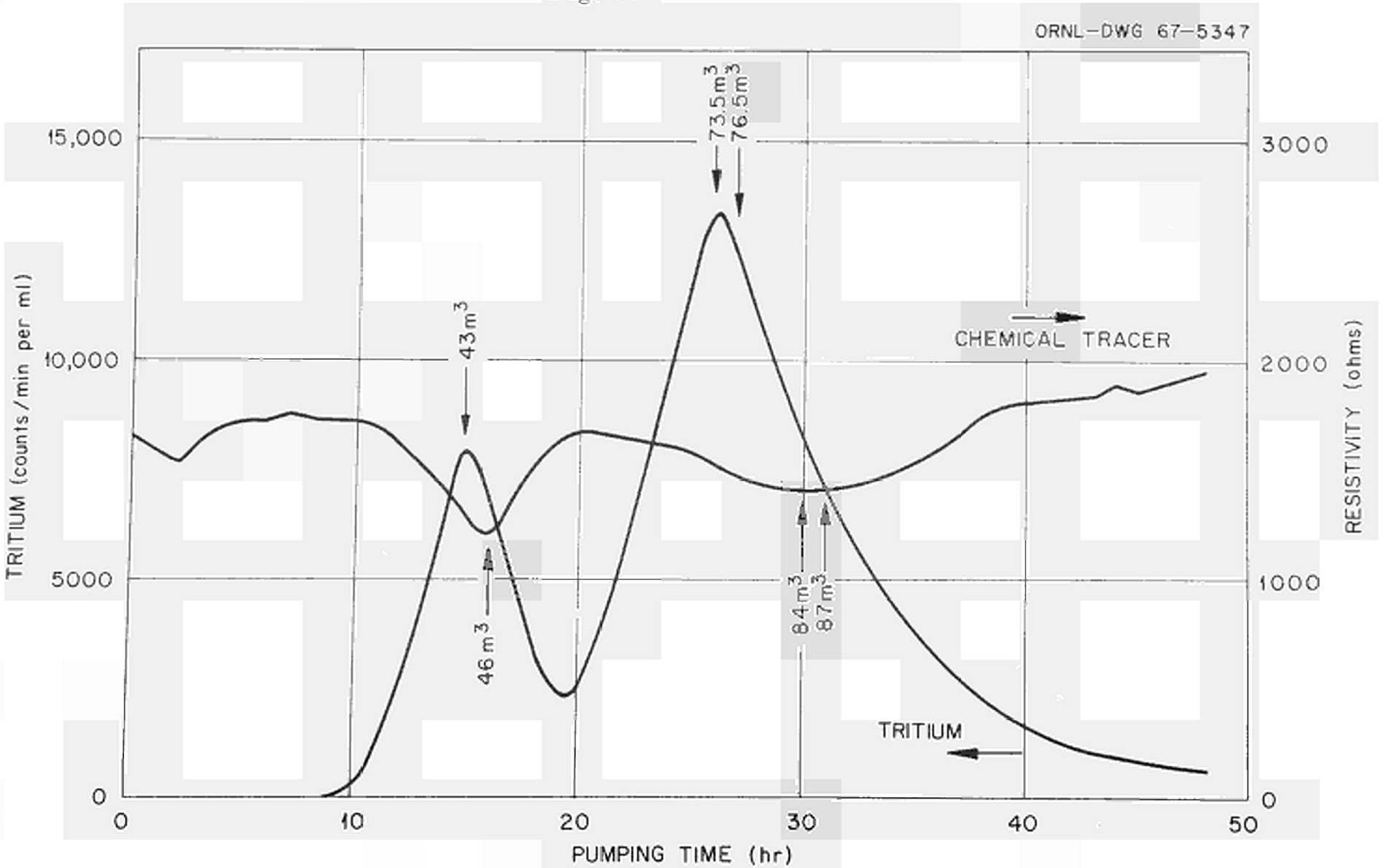
◎ P PUMPED WELL
SCREENED 4.5 TO 5.0 meters

○ B WELLS FOR INJECTION OF
BARRIER BUILDING CHEMICALS
SCREENED SURFACE TO 6 meters

● D DRY HOLES
NO SCREEN, DEPTH 6 meters

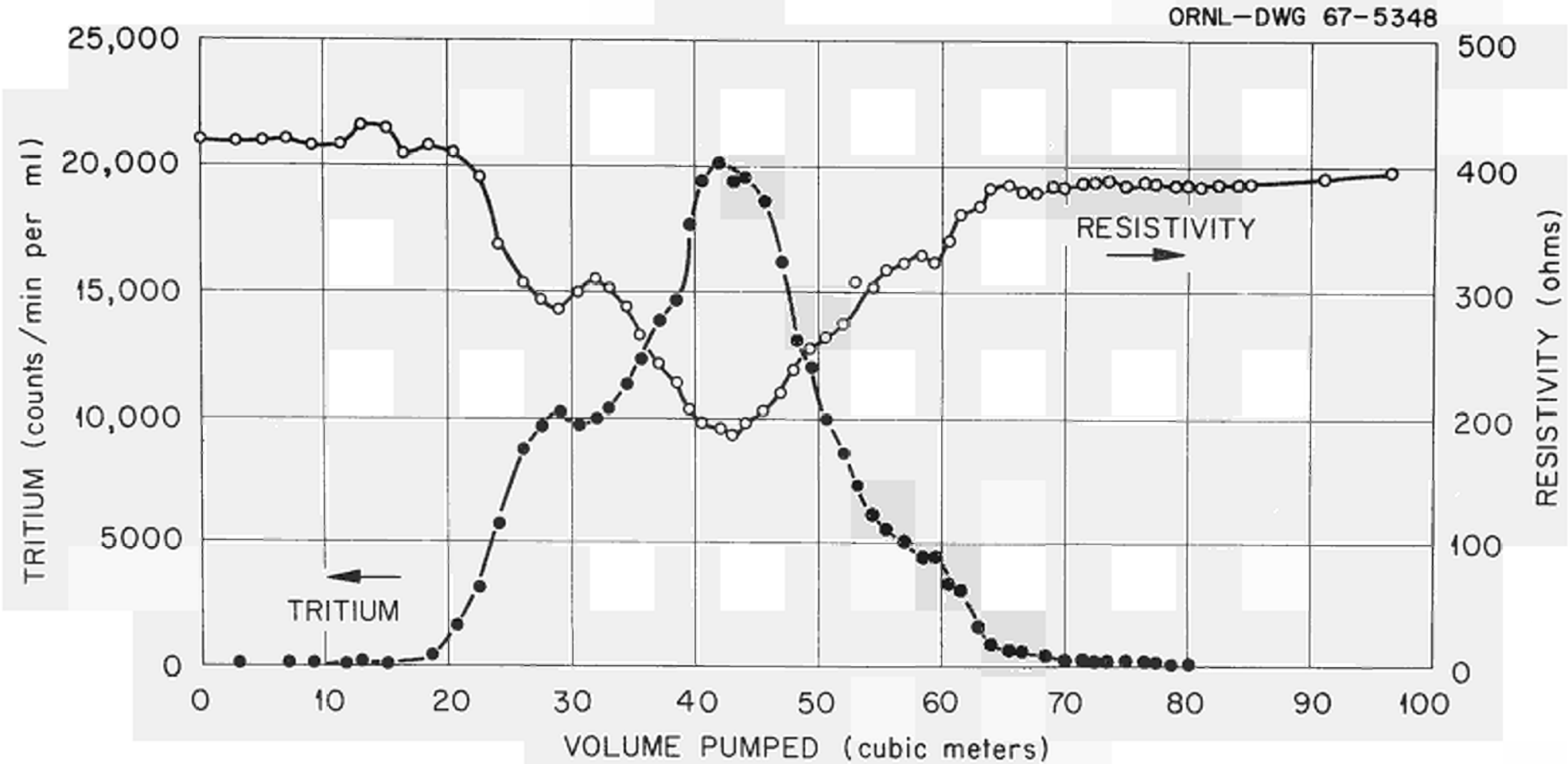
Wells and Dry Holes Used in Barrier Building Experiment

Fig. 21



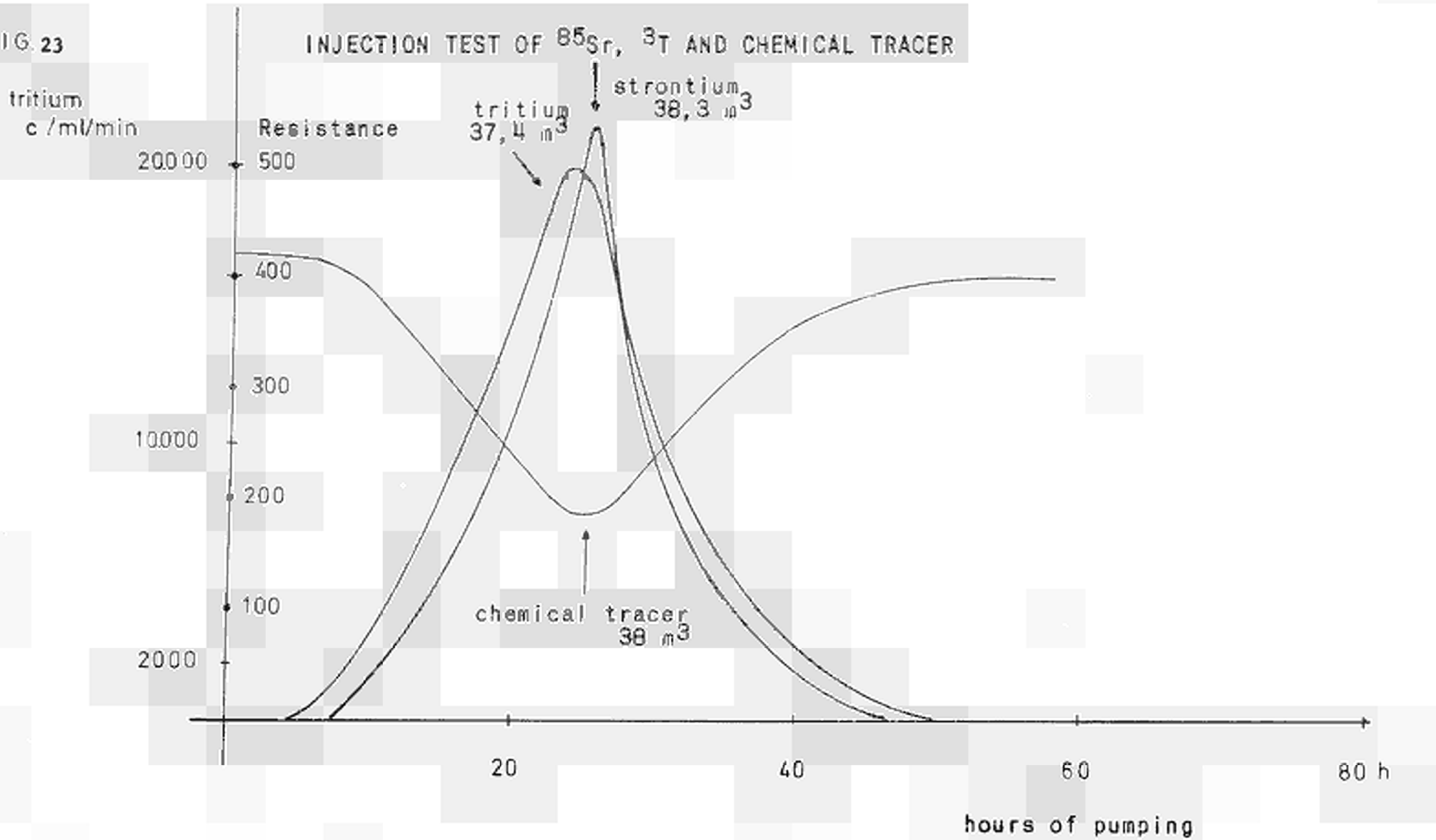
Tritium and Chemical Tracers, Preliminary Barrier Test, July, 1966.

Fig. 22



Tritium and Chemical Tracers; Preliminary Barrier Test, Nov 10 to 15, 1966.

FIG. 23


INJECTION TEST OF 85Sr, 3T AND CHEMICAL TRACER
tritium
c /ml/min
Resistance
20000
500
400
300
10000
200
100
2000
tritium
37,4 m3
strontium
38,3 m3
chemical tracer
38 m3
20
40
60
80 h
hours of pumping

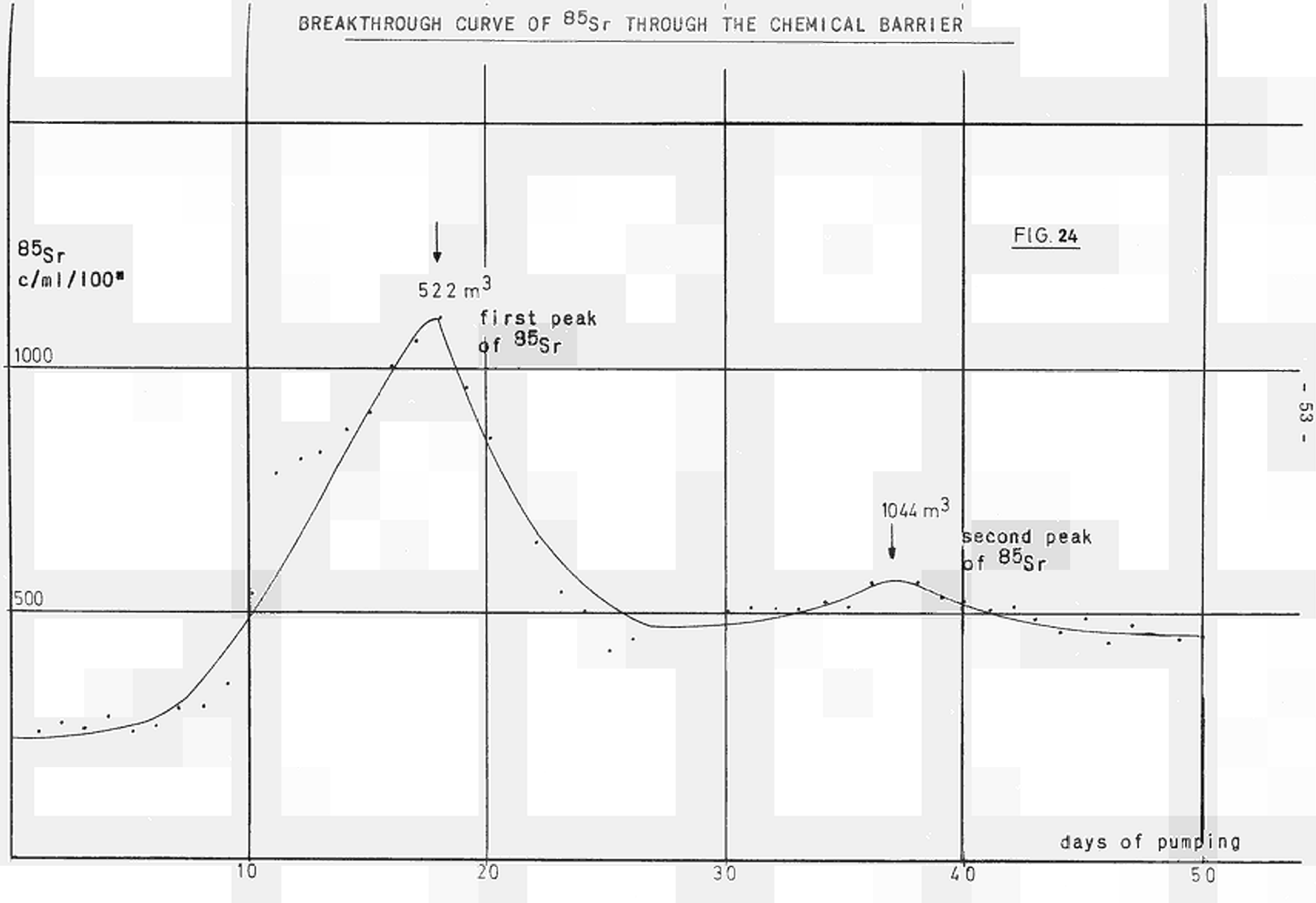

BREAKTHROUGH CURVE OF 85Sr THROUGH THE CHEMICAL BARRIER
85Sr c/ml/100*
1000
500
522 m3
first peak of 85Sr
1044 m3
second peak of 85Sr
10
20
30
40
50
days of pumping

FIG. 24

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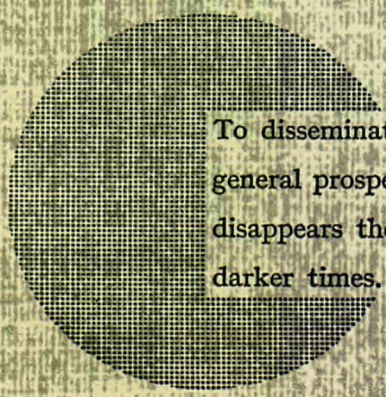

To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel

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